Enhanced generation of twin single-photon states via quantum interference in parametric down-conversion: Application to two-photon quantum photolithography

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Two-photon interferometric quantum photon lithography for light of wavelength λ is capable of beating the Rayleigh diffraction limit of resolution λ/4 to the level of λ/8. The required twin single-photon states $|1\rangle_a|1\rangle_b$, which are converted into maximally entangled states by a 50:50 beam splitter, can be generated from a nondegenerate parametric amplifier initially in vacuum states and with a weak pump field. Increasing the pump strength can slightly increase the production rate of the desired state and it will also increase the production of the twin two-photon states $|2\rangle_a|2\rangle_b$, which leads to an unwanted background term. In this paper we show that, assuming a weak pair coherent state as input to the amplifier, quantum interference can be used to quench the production of the $|2\rangle_a|2\rangle_b$ state and to enhance the production of the $|1\rangle_a|1\rangle_b$ state by almost sixfold.

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I. INTRODUCTION

In recent years, there has been some interest in the application of certain nonclassical states of a two-mode quantized light field to the problem of interferometric photolithography, also known as quantum lithography [1]. Photolithography has been the principal method by which the semiconductor industry transfers circuitry images onto a substrate. The resolution of images transferred by using classical light beams is restricted to the Rayleigh diffraction limit λ/4, λ being the wavelength of the light. Obviously, obtaining higher resolution to transfer smaller and smaller images with classical light requires shorter and shorter wavelength light. On the other hand, Boto *et al.* [1] have shown that if a two-mode maximally entangled state (MES) of the form

$$|N::0\rangle_{a,b}^{\Phi}=\frac{1}{\sqrt{2}}(|N\rangle_a|0\rangle_b+e^{i\Phi}|0\rangle_a|N\rangle_b) \tag{1.1}$$

can be produced, where we have adopted the notation of Kok, Lee, and Dowling [2], and if a substrate able to absorb only N photons at a time is available, the Rayleigh diffraction limit can be breached to $\lambda/4N$. Obstacles to overcome in implementing photolithography beyond the Rayleigh limit are those associated with the generation of the required MES for arbitrary N and the production of the required substrates. A number of schemes have been proposed for generating MES with various photon numbers N [2,3], but only the MES for $N=2$ are readily available from down-conversion followed by 50:50 beam splitting, as in the type of experiment performed some years ago by Hong, Ou, and Mandel [4]. With respect to substrates, there appears to be little hope, at least at this time, for the production of materials able to absorb *only* N photons for $N>2$. The case $N=2$ may be just possible if some way can be found to suppress independent single photon absorptions [5].

In connection with two-photon lithography, Nagasako *et al.* [6] have studied the use of a high-gain, single pass, parametric amplifier as an intense source of entangled light. In the limit of low gain, such an amplifier produces a stream of single-photon pairs, or twin single-photon states $|1\rangle_a|1\rangle_b$, which, when directed to the input ports of a 50:50 beam splitter, are converted into a MES of the general form $|2::0\rangle_{a,b}^{\Phi}$ [4]. The value of the phase Φ will depend on the internal construction of the beam splitter. When operated at high gain, the fringe visibility of the output is degraded, but is always at least 20%, and therefore the authors concluded that a high-gain parametric amplifier possesses great promise for certain applications in quantum optics, including quantum lithography. But in this regard, the two-photon deposition function on the substrate will contain an unwanted background term as a direct result of the high-gain parametric amplifier.

In this paper we reexamine the case of the low-gain parametric amplifier. With low gain, as already mentioned, the $|1\rangle_a|1\rangle_b$ state is generated and thus the desired MES $|2::0\rangle_{a,b}^{\Phi}$ is produced by subsequently directing the two photons to a 50:50 beam splitter. With slightly higher gain, the twin two-photon state $|2\rangle_a|2\rangle_b$ is generated [7], but a subsequent beam splitter *does not* produce a MES. With respect to lithography, it mainly gives rise to a background term in the deposition function. In most (if not all) of the discussions on using down-converted light from a parametric amplifier for the purposes of lithography, the input state has been assumed to be the vacuum. Here we consider the use of a weak pair coherent state as the input and show that, through quantum interference, it is possible to *increase* the production of the $|1\rangle_a|1\rangle_b$ state and at the same time *decrease* the production of the $|2\rangle_a|2\rangle_b$ state. Essentially, it is the quantum interference between the action of the parametric amplifier and the input pair coherent state that is responsible for this behavior.

Recently, there has been much activity in connection with the generation of certain kinds of nonclassical photonic states via quantum interference in both single-mode and two-mode parametric amplifiers [8–10]. This work is largely based on a proposal made some years ago (1974) by Stoler [11] for the production of antibunched light, a proposal that was not realized experimentally until almost 20 years later [12]. Most of the work cited in Refs. [8–10] is concerned with the removal or enhancement, via destructive or con-

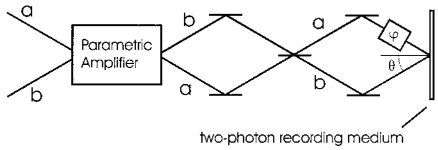

FIG. 1. Schematic for two-photon lithography using a parametric amplifier. The beam splitter is assumed to be 50:50. The box in the upper beam on the right represents the relative phase shift $\varphi = 2\pi x/\lambda$, where x is the lateral distance along the medium. The beams are assumed the incident on the substrate at the grazing angle where $\theta \to \pi/2$.

structive quantum interference, respectively, of the two-photon term that appears from the first-order time-dependent perturbation expansion, valid for short interaction times, with a coherent state input field. The present work uses quantum interference in a two-mode system with an input weak pair coherent state to suppress the production of the unwanted state $|2\rangle_a|2\rangle_b$ by destructive interference involving the second order of the perturbative expansion of the time-evolution operator for the parametric amplifier and to simultaneously enhance the production of the desired $|1\rangle_a|1\rangle_b$ state by constructive interference in the first-order term. The state generated by this method is, in fact, a weak form of the two-mode squeezed pair coherent state discussed by this author some years ago [13].

The paper is structured as follows. In Sec. II we review the ideas behind quantum photolithography with special attention given to the two-photon case with light from a low-gain parametric amplifier with input vacuum states. In Sec. III we study the interference effects arising from an input pair coherent state. In Sec. IV we conclude the paper with some brief remarks.

II. TWO-PHOTON QUANTUM PHOTOLITHOGRAPHY

Figure 1 is a schematic for two-photon lithography using a parametric amplifier. The parametric amplifier is described by the interaction Hamiltonian

$$\hat{H}_I = i\hbar\kappa(\hat{a}^\dagger\hat{b}^\dagger - \hat{a}\hat{b}). \tag{2.1}$$

The parameter κ is assumed to be real and is proportional to the second-order nonlinear susceptibility of the crystal and the strength of the pump field, the field assumed to be classical. We suppose here that the input states are vacuum states $|0\rangle_a|0\rangle_b$. As we are interested only in weak pump fields, we use perturbation theory to obtain the time-evolved state, to second order in time, as

$$\begin{aligned}|\Phi(t)\rangle &\approx [1 - i\hat{H}_I t/\hbar + \tfrac{1}{2}(-i\hat{H}_I t/\hbar)^2 + \cdots]|0\rangle_a|0\rangle_b \\ &= (1-\eta^2/2)|0\rangle_a|0\rangle_b + \eta|1\rangle_a|1\rangle_b \\ &\quad + \eta^2|2\rangle_a|2\rangle_b,\end{aligned} \tag{2.2}$$

where we have set $\eta = \kappa t$. Let us suppose now that the pump field is weak enough to ignore the second-order term so that we have, to first order in η,

$$|\Phi(t)\rangle \approx (1-\eta^2/2)|0\rangle_a|0\rangle_b + \eta|1\rangle_a|1\rangle_b. \tag{2.3}$$

The beam splitter generates the state

$$\begin{aligned}|\Phi_{\mathrm{BS}}\rangle &= (1-\eta^2/2)|0\rangle_a|0\rangle_b + i\eta(|2\rangle_a|0\rangle_b + |0\rangle_a|2\rangle_b)/\sqrt{2} \\ &= (1-\eta^2/2)|0\rangle_a|0\rangle_b + i\eta|2::0\rangle^0_{a,b},\end{aligned} \tag{2.4}$$

where $|2::0\rangle^0_{a,b}$ is the maximally entangled two-photon state, using the notation of Eq. (1.1). We have assumed the beam splitter to be 50:50 and to be described by the transformation $\hat{U}_{\mathrm{BS}} = \exp[i\pi(\hat{a}^\dagger\hat{b} + \hat{b}^\dagger\hat{a})/4]$ (Ref. [14]). This represents a particular layering of the dielectrics used to construct the beam splitter.

We next assume that the output beams of the beam splitter are directed to a two-photon recording substrate as pictured in Fig. 1. The block in the upper beam represents the relative phase shift $\varphi = 2\pi x/\lambda$ between the two beams on the surface of the substrate, where, again, λ is the wavelength of the light and x is the lateral distance along the substrate. We further assume that the beams are incident upon the surface at the grazing angle, i.e., that in Fig. 1 the angle $\theta \to \pi/2$. The dosing operator for the two-photon substrate is then given by $\hat{\delta}_2 = \hat{e}^{\dagger 2}\hat{e}^2/2!$ where $\hat{e} = \hat{a} + \hat{b}$ is the superposition mode operator. Representing the relative phase shift, as a phase shift in the upper beam, by the operator $\exp(i\varphi\hat{a}^\dagger\hat{a})$, then from Eq. (2.4) the state on the substrate is

$$|\Phi_{\mathrm{sub}}\rangle = (1-\eta^2/2)|0\rangle_a|0\rangle_b + i\eta e^{2i\varphi}|2::0\rangle^{-2\varphi}_{a,b}. \tag{2.5}$$

The relevant quantity for quantum photolithography is the deposition function

$$\Delta_{2,\gamma} = \langle\Phi_{\mathrm{sub}}|\hat{\delta}_2|\Phi_{\mathrm{sub}}\rangle = \eta^2[1+\cos(2\varphi)]. \tag{2.6}$$

Note that the vacuum term makes no contribution. At the grazing limit, the spatial oscillation represents a resolution of $\Delta x = \lambda/8$.

But the parameter η is small and thus the rate of two-photon deposition will be low. We may increase η by increasing the strength of the pump field or increasing the interaction time. But this means that the four-photon term of Eq. (2.2) must now be retained. In this case, the state after the beam splitter will be

$$\begin{aligned}|\Phi_{\mathrm{BS}}\rangle &= (1-\eta^2/2)|0\rangle_a|0\rangle_b + i\eta(|2\rangle_a|0\rangle_b + |0\rangle_a|2\rangle_b)/\sqrt{2} \\ &\quad - \eta^2\left[\sqrt{\tfrac{3}{8}}(|4\rangle_a|0\rangle_b + |0\rangle_a|4\rangle_b) + \tfrac{1}{2}|2\rangle_a|2\rangle_b\right].\end{aligned} \tag{2.7}$$

Recalculating the deposition function we find

$$\Delta_{2,\gamma} = (\eta^2 + 3\eta^4)[1+\cos(2\varphi)] + 4\eta^4. \tag{2.8}$$

The deposition rate is slightly enhanced, but only to the forth order in η, and at the same time, an unwanted background term $4\eta^4$ is present. Increasing the pump field brings about an even larger background term as is clear from the work of Nagasako *et al.* [6].

In the next section, we show how a different choice of initial state for the parametric amplifier, namely, a pair coherent, state allows for the significant enhancement of the deposition rate and for the quenching of the terms giving rise to the background, all by quantum interference.

III. INTERFERENCE WITH A PAIR COHERENT STATE

The pair coherent states $|\zeta,q\rangle$ are defined as eigenstates of the pair annihilation operator $\hat{a}\hat{b}$ and the difference operator $\hat{\Delta}=\hat{a}^{\dagger}\hat{a}-\hat{b}^{\dagger}\hat{b}$ according to [15]

$$\hat{a}\hat{b}|\zeta,q\rangle=\zeta|\zeta,q\rangle, \quad \hat{\Delta}|\zeta,q\rangle=q|\zeta,q\rangle, \tag{3.1}$$

where ζ is a complex number unrestricted over the complex plane, and q, known as the degeneracy parameter, is an integer. We are interested only in the degenerate case where $q=0$, and thus the solution to Eqs. (3.1) is

$$|\zeta,0\rangle\equiv|\zeta\rangle=\mathcal{N}\sum_{n=0}^{\infty}\frac{\zeta^n}{n!}|n\rangle_a|n\rangle_b, \quad \mathcal{N}=\left(\sum_{n=0}^{\infty}\frac{|\zeta|^{2n}}{(n!)^2}\right)^{-1/2}, \tag{3.2}$$

where we have dropped the q label. A pair coherent state can be generated via the competition between a two-photon parametric process and a two-photon absorption process [15], or by a nondegenerate parametric oscillator [16].

For a weak pair coherent state, we have, to second order in ζ,

$$|\zeta\rangle\approx\mathcal{N}\Big(|0\rangle_a|0\rangle_b+\zeta|1\rangle_a|1\rangle_b+\frac{\zeta^2}{2}|2\rangle_a|2\rangle_b,$$
$$\mathcal{N}\approx(1+|\zeta|^2+|\zeta|^4/4+\cdots)^{-1/2}. \tag{3.3}$$

With such a state as the input to the parametric amplifier, the state vector just before the beam splitter is

$$|\Phi\rangle\approx\mathcal{N}[(1-\zeta\eta-\eta^2/2)|0\rangle_a|0\rangle_b+(\eta+\zeta)|1\rangle_a|1\rangle_b$$
$$+(\eta^2+2\zeta\eta+\zeta^2/2)|2\rangle_a|2\rangle_b]. \tag{3.4}$$

We suppose now that ζ is chosen such that the coefficient of $|2\rangle_a|2\rangle_b$ vanishes, i.e., ζ is a root of $\zeta^2+4\zeta\eta+2\eta^2=0$. The roots are easily found to be $\zeta_{\pm}=(-2\pm\sqrt{2})\eta$. As we wish to maximize the coefficient of $|1\rangle_a|1\rangle_b$, we choose the root $\zeta_{-}=-(2+\sqrt{2})\eta$. The deposition function on the substrate will then be

$$\Delta_{2,\gamma}=(1+\sqrt{2})^2\eta^2[1+\cos(2\varphi)]\approx5.828\eta^2[1+\cos(2\varphi)]. \tag{3.5}$$

Thus, the deposition rate is almost six times that for an initial vacuum state and no background term is present, both features the result of quantum interference.

IV. CONCLUSIONS

In this short paper, we have shown how it should be possible to use quantum interference effects, with a weak pair coherent state as the input, to suppress the production of twin two-photon states that lead to background terms and at the same time enhance the production of the twin single-photon states that ultimately, through beam splitting, can be converted into two-photon maximally entangled states. Such states can be used for photolithography as discussed above, but they could also be used for two-photon interferometry where the Heisenberg limit of sensitivity for measuring phase shifts, in this case $\Delta\varphi=1/2$ [it is $\Delta\varphi=1/N$ for the N-photon maximally entangled state of Eq. (1.1)], can be attained. Finally, it is perhaps worthwhile to out that for the interference effect to work in the described manner, the input state to the parametric amplifier needs to be a superposition of the form $\Sigma_n c_n|n\rangle_a|n\rangle_b$, i.e., a superposition of twin Fock states. Of course, a two-mode squeezed vacuum state is of this form, but it is easy to see that the condition under which the $|2\rangle_a|2\rangle_b$ state is removed by interference is precisely the condition that *all* the states, save the vacuum, will be removed by interference. This is just the inverse of the transformation that generates the two-mode squeezed vacuum state from the vacuum in the first place. If we should choose instead uncorrelated ordinary coherent states for the two modes, unwanted states, such as $|2\rangle_a|0\rangle_b$ and $|0\rangle_a|2\rangle_b$, will be generated. So we have studied the case where the initial state is a degenerate weak pair coherent state. Of course, such a state may not be easy to generate in its own right as is highly nonclassical. But it must be kept in mind that the two-photon maximally entangled state too is quite nonclassical and it should be no surprise that generating an enhanced form of one type of nonclassical light may require the manipulation of some other type.

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[1] A. Boto, P. Kok, D. S. Abrams, S. L. Braunstein, C. P. Williams, and J. P. Dowling, Phys. Rev. Lett. **85**, 2733 (2000).
[2] P. Kok, H. Lee, and J. P. Dowling, Phys. Rev. A **65**, 052104 (2002).
[3] C. C. Gerry, Phys. Rev. A **61**, 043811 (2000); C. C. Gerry and R. A. Campos, *ibid.* **64**, 063814 (2001); H. Lee, P. Kok, N. Cerf, and J. P. Dowling, *ibid.* **65**, 030101(R) (2002); C. C. Gerry and A. Benmoussa, *ibid.* **65**, 033822 (2002); J. Fiurášek, *ibid.* **65**, 053818 (2002); X. Zou, K. Pahlke, and W. Mathis, *ibid.* **66**, 014102 (2002); C. C. Gerry, A. Benmoussa, and R. A. Campos, *ibid.* **66**, 013804 (2002); C. C. Gerry and A. Benmoussa, *ibid.* **66**, 033804 (2002).
[4] C. K. Hong, Z. Y. Ou, and L. Mandel, Phys. Rev. Lett. **59**, 2044 (1987).
[5] See J. Javanainen and P. L. Gould, Phys. Rev. A **41**, 5088 (1990).
[6] E. M. Nagasako, S. J. Bentley, R. W. Boyd, and G. S. Agarwal, Phys. Rev. A **64**, 043802 (2001).

[7] Z. Y. Ou, J.-K. Rhee, and L. J. Wang, Phys. Rev. Lett. **83**, 959 (1999).
[8] Y. J. Lu and Z. Y. Ou, Phys. Rev. Lett. **88**, 023601 (2002).
[9] K. J. Resch, J. S. Lundeen, and A. M. Steinberg, Phys. Rev. Lett. **88**, 113601 (2002).
[10] R.-H. Xie, Phys. Rev. A **65**, 055801 (2002).
[11] D. Stoler, Phys. Rev. Lett. **33**, 1379 (1974).
[12] M. Koashi, K. T. Hirano, and M. Matsuoka, Phys. Rev. Lett. **71**, 1164 (1993).
[13] C. C. Gerry, J. Mod. Opt. **42**, 585 (1995).
[14] B. Yurke, S. L. McCall, and J. R. Klauder, Phys. Rev. A **33**, 4033 (1985); R. A. Campos, B. E. A. Saleh, and M. C. Teich, *ibid.* **40**, 1371 (1989).
[15] G. S. Agarwal, Phys. Rev. Lett. **57**, 827 (1986); J. Opt. Soc. Am. B **5**, 1940 (1988).
[16] M. D. Reid and L. Krippner, Phys. Rev. A **47**, 552 (1993); A. Gilchrist and W. J. Munro, J. Opt. B: Quantum Semiclassical Opt. **2**, 47 (2000).