# Heisenberg-limited interferometry and photolithography with nonlinear four-wave mixing

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Maximally entangled photonic states of a two-mode field have applications in Heisenberg-limit interferometry and in photolithography where they may be used to transfer images with resolutions exceeding the Rayleigh diffraction limit. In a recent paper by one of us [C. C. Gerry, Phys. Rev. A **61**, 043811 (2000)] it was shown that a nonlinear four-wave mixer could produce the requisite states for input states containing only even photon numbers. For superpositions of even number states the output is just a superposition of maximally entangled even states. In the present paper we extend the earlier work to consider both even coherent states and squeezed vacuum states as inputs and study their applications to interferometry and lithography.

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Currently there is much interest in the generation of the so-called maximally entangled states (MES) of a two-mode quantized electromagnetic field. For a total of $n$ photons in a two-mode field, with the modes labeled $a$ and $b$, the MES have the generic form

$$\frac{1}{\sqrt{2}}(|n\rangle_a|0\rangle_b+e^{i\vartheta}|0\rangle_a|n\rangle_b). \qquad (1)$$

These states are of interest because of at least two possible practical applications: they would allow optical interferometry to be performed at the Heisenberg limit of phase uncertainty [1], $\Delta\varphi_H=1/n$, the greatest level of sensitivity allowed by quantum theory [2], and would enable interferometric photolithography beyond the Rayleigh diffraction limit [3]. However the prospect of generating the required states of Eq. (1) is nontrivial. In a recent paper [4], one of the present authors proposed a method involving the use of a nonlinear four-wave mixer (NFWM), a device previously discussed by Yurke and Stoler [5]. The interaction involves the usual four-wave mixing term competing with a Kerr-like term. In [5] it was shown that under certain operating conditions the device would act as an even-odd filter with respect to photon number. But in [4] it was shown that under different operating conditions it could produce the MES of the form of Eq. (1) as long as $n$ were even. It turns out that through the introduction of the Schwinger realization of the angular momentum operators in terms of two sets of Bose operators, the interaction involved has the same mathematical form as the nonlinear spin interaction discussed by Mølmer and Sørensen [6] for generating MES for the internal states of a system of N two-level hot trapped ions, a scheme which has since been realized experimentally [7]. For the greatest sensitivity in interferometry $n$ should be as large as possible but the required input number states $|n\rangle$, especially for high $n$, are generally not available. Thus in [4] a superposition of even number states, in fact the even coherent (Schrödinger cat) states, states that could be obtained from another NFWM operated as described in [5], were studied and shown to reach the Heisenberg limit in terms of the average photon number $\bar{n}$ of the even coherent state, i.e., $\Delta\varphi=1/\bar{n}$. In the present paper we reexamine the NFWM device for the input even coherent states, extending our considerations to the case of the squeezed vacuum as input states, and then apply our output states to interferometry and quantum photolithography.

In Fig. 1, we illustrate our prototype for interferometric measurements with a Mach-Zehnder interferometer (MZI) device except where the first beam splitter has been replaced by an NFWM. Assuming the two modes are degenerate in frequency, the Hamiltonian of the NFWM is given by

$$H=\hbar\omega(a^\dagger a+b^\dagger b)+\hbar\frac{\Omega}{4}(a^\dagger b+ab^\dagger)^2. \qquad (2)$$

Upon expansion we have

$$H=\hbar\omega(a^\dagger a+b^\dagger b)+\hbar\frac{\Omega}{4}(a^{\dagger 2}b^2+a^2b^{\dagger 2}+2a^\dagger ab^\dagger b + a^\dagger a+b^\dagger b). \qquad (3)$$

The first two interaction terms are the usual four-wave mixing interactions, the third a cross-Kerr interaction, and the last two terms give rise to a frequency shift for the two modes. Henceforth we shall work in the interaction picture where the Hamiltonian becomes

$$H_{\rm IP}=\hbar\frac{\Omega}{4}(a^\dagger b+ab^\dagger)^2. \qquad (4)$$

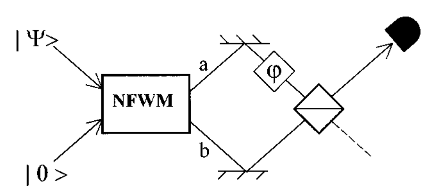


FIG. 1. Modified Mach-Zehnder interferometer where the first beam splitter has been replaced by a nonlinear four-wave mixer operating under the conditions required to produce maximally entangled states for an even input state $|\Psi\rangle$. Detection is performed only on the output $b$ mode.

 

The connection to a nonlinear spin interaction arises through introducing the Schwinger operators [8]

$$J_1=(a^\dagger b+ab^\dagger)/2,$$
$$J_2=(a^\dagger b-ab^\dagger)/2i,$$
$$J_3=(a^\dagger a-b^\dagger b)/2 \qquad (5)$$

satisfying the angular momentum algebra $[J_i,J_j]=i\varepsilon_{ijk}J_k$, such that Eq. (4) may be written as $H_{\mathrm{IP}}=\hbar\Omega J_1^2$.

For the input state $|2m\rangle_a|0\rangle_b$, $m=0,1,2,\ldots,$ where we have taken the number states in the $a$ mode to be even, and for the interaction time $T_{\mathrm{MES}}=\pi/2\Omega$, we obtain [4]

$$|2m\rangle_{\mathrm{MES}}=\exp(-iH_{\mathrm{IP}}T_{\mathrm{MES}}/\hbar)|2m\rangle_a|0\rangle_b$$
$$=\frac{1}{\sqrt{2}}(|2m\rangle_a|0\rangle_b+e^{-\Phi_{2m}}|0\rangle_a|2m\rangle_b), \qquad (6)$$

where $\Phi_{2m}=(2m+1)\pi/2$. We let the phase-shift operation in the upper arm of the MZI of Fig. 1 be represented by the operator $U(\varphi)=\exp(i\varphi a^\dagger a)$ and we let the beam splitter (assumed 50:50) be represented by $U_{\mathrm{BS}}=\exp[i\pi(a^\dagger b+ab^\dagger)/4]=\exp(i\pi J_1/2)$ [9]. The latter operator represents a particular choice of internal phases of the beam splitter, chosen for convenience, the final results for the phase uncertainty being independent of these internal phases. Thus the output of the MZI is

$$|\mathrm{out}\rangle_{\mathrm{MZI}}=U_{\mathrm{BS}}U(\varphi)|2m\rangle_{\mathrm{MES}}$$
$$=U_{\mathrm{BS}}\frac{1}{\sqrt{2}}(e^{2im\varphi}|2m\rangle_n|0\rangle_b+e^{-i\Phi_{2m}}|0\rangle_a|2m\rangle_b). \qquad (7)$$

In a typical MZI experiment involving only passive beam splitters, one generally measures the difference in the photon numbers in the output beams of the second beam splitter, essentially the expectation value of the operator $J_3$ of Eq. (4) [10]. But for the states of Eq. (6) we have $\langle J_3\rangle=0$. To circumvent this problem, Bollinger *et al.* [1] suggested measuring the parity operator of one of the output modes. Choosing the $b$ mode, this operator may be written as

$$O=(-1)^{b^\dagger b}=\exp[i\pi(J_0-J_3)], \qquad (8)$$

where $J_0=(a^\dagger a+b^\dagger b)/2$ is the total number of photons in both modes. $J_0$ commutes with all the angular momentum operators of Eq. (4). Detecting the observable $O$ is equivalent to measuring the number of photons $n_b$ emerging in the $b$ mode and assigning the measurement the value $(-1)^{n_b}$. This in turn is equivalent to measuring all the moments of the number operator $b^\dagger b$. For the state of Eq. (6) we have

$$\langle O\rangle={}_{\mathrm{MZI}}\langle\mathrm{out}|\exp[i\pi(J_0-J_3)]|\mathrm{out}\rangle_{\mathrm{MZI}}$$
$$=(-1)^m\cos(2m\varphi+\Phi_{2m}), \qquad (9)$$

where we have used the results

$$\exp(-i\pi J_1/2)J_3\exp(i\pi J_1/2)=J_2,$$
$$\exp(i\pi J_2)|n\rangle_a|l\rangle_b=(-1)^n|l\rangle_a|n\rangle_b. \qquad (10)$$

The phase uncertainty is then given by

$$\Delta\varphi=\Delta O\bigg/\left|\frac{\partial\langle O\rangle}{\partial\varphi}\right|, \qquad (11)$$

where, since $O^2=1$, $\Delta O=\sqrt{1-\langle O\rangle^2}=\sin(2m\varphi+\Phi_{2m})$. From this it easily follows that $\Delta\varphi=1/(2m)$, exactly the Heisenberg limit.

We now consider as input to the NFWM the more general state consisting of a superposition of only the even number photon states which we write as

$$|\psi\rangle=\sum_{m=0}^{\infty}C_{2m}|2m\rangle_a, \qquad (12)$$

and for which the average photon number is

$$\bar{n}=\sum_{m=0}^{\infty}(2m)|C_{2m}|^2.$$

The output of the NFWM is now

$$|\mathrm{out}\rangle_{\mathrm{NFWM}}=\sum_{m=0}^{\infty}C_{2m}|2m\rangle_{\mathrm{MES}}$$
$$=\frac{1}{\sqrt{2}}\sum_{m=0}^{\infty}C_{2m}[|2m\rangle_a|0\rangle_b+e^{-i\Phi_{2m}}|0\rangle_a|2m\rangle_b], \qquad (13)$$

and the expectation value of the operator $O$ is then

$$\langle O\rangle=\sum_{m=0}^{\infty}|C_{2m}|^2(-1)^m\cos(2m\varphi+\Phi_{2m}). \qquad (14)$$

We consider as inputs two kinds of even number photon states as mentioned above: the even coherent states and the squeezed vacuum states. For the former, the state is denoted $|z\rangle_{\mathrm{ECS}}$ and the coefficients are [11]

$$C_{2m}=[\cosh(|z|)]^{-1/2}\frac{z^m}{\sqrt{(2m)!}}. \qquad (15)$$

The average photon number is $\bar{n}_{\mathrm{ECS}}=|z|\tanh(|z|)$. Recall that the photon number probability distribution is similar to that of the Poisson distribution but with the important difference that all the odd number states have zero probabilities. The peak of the distribution is near $\bar{n}_{\mathrm{ECS}}$. The parameter $z$ is complex and $0\leqslant|z|<\infty$. The even coherent state may be written as a superposition of the usual coherent states as

$$|z\rangle_{\mathrm{ECS}}=\frac{1}{\sqrt{2}}(1+e^{-2|\alpha|^2})(|\alpha\rangle+|-\alpha\rangle), \quad z=\alpha^2, \qquad (16)$$

which is a form of Schrödinger cat state [12]. Note that it satisfies the eigenvalue relation $a^2|z\rangle_{\mathrm{MES}}=z|z\rangle_{\mathrm{MES}}$. The output state in this case may be written as

$$|\text{out}\rangle_{\text{NFWM}} = \frac{1}{\sqrt{2}}(|z\rangle_{\text{ECS},a}|0\rangle_b - i|0\rangle_a|-z\rangle_{\text{ECS},b}), \quad (17)$$

an entanglement of vacuum and even coherent states.

For the squeezed vacuum state [13], which we denote in the customary way as $|\xi\rangle_{\text{SV}}$, the coefficients are

$$C_{2m} = (\cosh r)^{-1/2}(-1)^m \frac{\sqrt{(2m)!}}{2^m m!}(e^{i\theta}\tanh r)^m, \quad (18)$$

where $\xi = e^{i\theta}\tanh r$, $0 \leqslant \theta < 2\pi$, $0 \leqslant r < \infty$, $r$ being the squeeze parameter. The average photon number for this state is $\bar{n}_{\text{SV}} = \sinh^2 r$. The corresponding photon number probability distribution is similar to that of thermal light except that, again, all odd states are missing and, of course, the squeezed vacuum states are pure. In this case we have

$$|\text{out}\rangle_{\text{NFWM}} = \frac{1}{\sqrt{2}}(|\xi\rangle_{\text{SV},a}|0\rangle_b - i|0\rangle_a|-\xi\rangle_{\text{SV},b}). \quad (19)$$

As it turns out, there exist closed form expressions for $\langle O\rangle$ for each of these states. For the even coherent states

$$\langle O\rangle = -\sinh(|z\rangle\cos\varphi)\sin(|z|\sin\varphi)/\cosh|z|, \quad (20a)$$

and for the squeezed vacuum states

$$\langle O\rangle = \sin(\Lambda/2)/(1+\sinh^2(2r)\sin^2(\varphi))^{1/4},$$

$$\Lambda = \tan^{-1}\left(\frac{-\sinh^2 r\sin(2\varphi)}{1+2\sinh^2 r\sin^2\varphi}\right). \quad (20b)$$

Notice that the results depend on the phase $\varphi$ [14]. For a balanced MZI $\varphi = 0$. Under this condition it is easy to show that $\Delta\varphi = 1/\bar{n}$, $\bar{n}$ being either $\bar{n}_{\text{ECS}}$ or $\bar{n}_{\text{SV}}$. Thus for a balanced interferometer we obtain sensitivities at the Heisenberg limit in terms of the average photon number of the input even state. This procedure of using an even coherent state as an input, a state that is perhaps a challenge to generate in its own right, makes it possible to approach the Heisenberg limit of sensitivity without the need for generating an even number state for the input to the NFWM. But if the MZI is not balanced, i.e., if $\varphi \neq 0$, the phase uncertainty will generally deviate from the Heisenberg limit but may still be improved over the corresponding standard quantum limit.

In Figs. 2 and 3 we plot the phase uncertainty versus average photon number for the input even coherent state. Figure 2(a) is for $\varphi = \pi/45$ while Fig. 2(b) is for $\varphi = \pi/18$. The exact Heisenberg-limit curve $\Delta\varphi_H = 1/\bar{n}$ is also shown. Aside from the periodic spikes the phase uncertainties closely follow the Heisenberg-limit curve, at least for small $\varphi$. By way of comparison, the sensitivity obtained from a standard MZI with a coherent state of average photon number $\bar{n}$ at one of the inputs, is $\Delta\varphi = 1/(1/\sqrt{\bar{n}}\sin\varphi)$ which obviously has the greatest sensitivity for $\varphi = \pi/2$, i.e., for an MZI having a phase difference of $\pi/2$ between the arms. Of course, with the insertion of a $\pi/2$ phase shifter it is possible to effectively rebalance the interferometer. In any case the

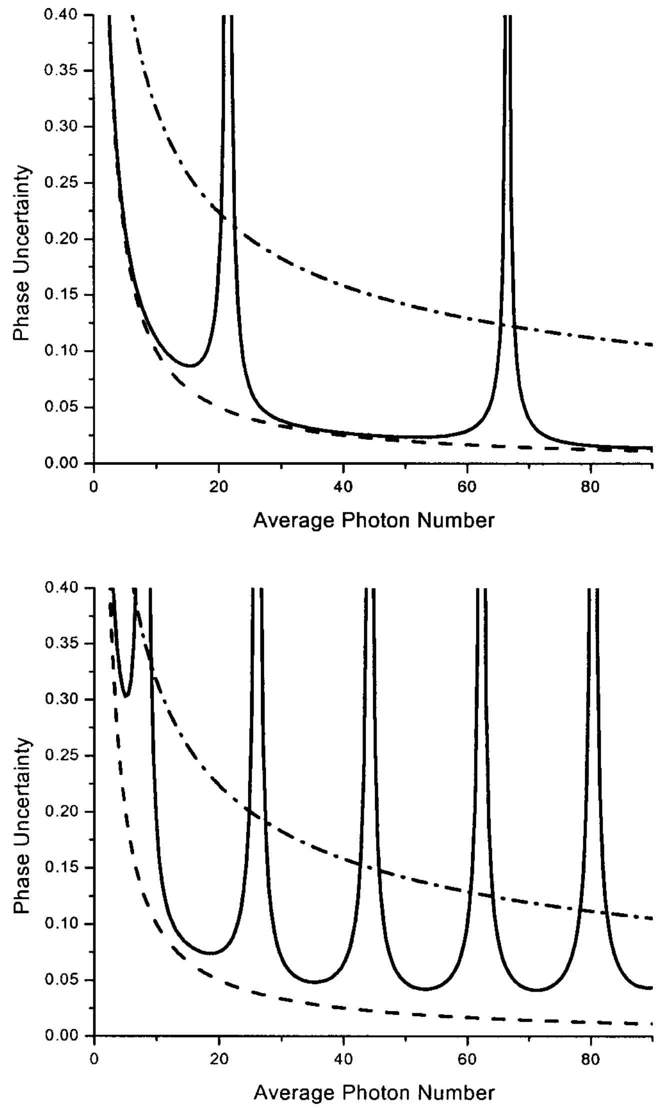


FIG. 2. Phase uncertainty $\Delta\varphi$ versus average photon number $\bar{n}$ for the input even coherent states (solid line) for the phase difference (a) $\varphi = \pi/45$ and (b) $\varphi = \pi/18$. The dashed lines represent the Heisenberg limit $1/\bar{n}$ while the dot-dashed lines represent the standard quantum limit $1/\sqrt{\bar{n}}$.

optimal level of sensitivity available with ''classical'' light, is the standard quantum limit (SQL), $\Delta\varphi_{\text{SQL}} = 1/\sqrt{\bar{n}}$. We include the SQL phase uncertainty in our figures for the sake of comparison. We notice that in Fig. 2, even when $\varphi$ is large enough to cause significant deviations from the Heisenberg limit, the phase uncertainty is still lower than the standard quantum limit.

But for the squeezed vacuum input the situation is different. For $\varphi$ only slightly different than zero, there are significant deviations from the Heisenberg limit as we show in Fig. 3 for $\varphi = \pi/90$. It is evident that the phase uncertainty in this case becomes markedly noisy as the average photon number increases, even exceeding that of the standard quantum limit. The difference in the results obtained for these two types of input states lies in the nature of the respective photon probability distributions. For the even coherent states, even though all odd number states are missing, the photon number

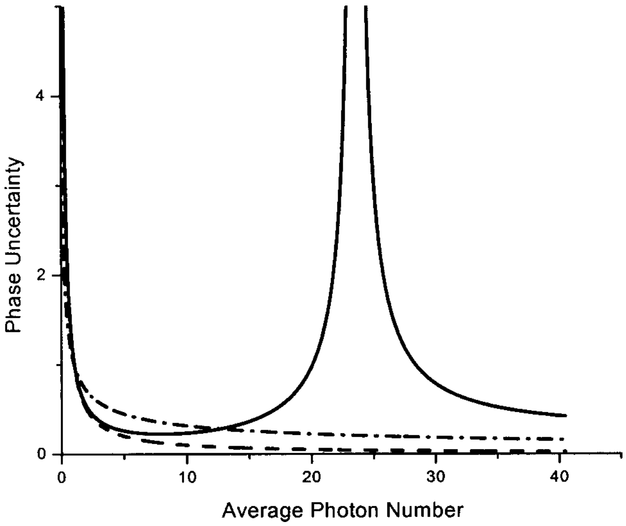


FIG. 3. Same as Fig. 2 but for the input squeezed vacuum states and only for $\varphi = \pi/90$.

distribution is peaked near the average photon number $\overline{n}_{\mathrm{ECS}}$. For the squeezed vacuum the distribution is thermal-like and for increasing $\overline{n}_{\mathrm{SV}}$ becomes extremely flat. This in turn means that a wide range of number states are contributing to the expectation value of the operator $O$. Clearly it is not enough just to have a superposition of even number states but rather there needs to be a least some degree of localization in the photon number distribution. Ideally it appears that the distribution should be sub-Poissonian. Recall that for an input even number state (a number state being the ultimate sub-Poissonian state) the phase uncertainty is exactly the Heisenberg limit with no dependence on the phase $\varphi$. But as is well known, the squeezed vacuum states are super-Poissonian, just the opposite of the ideal input states.

Finally, we consider applications of these states to photolithography. Diffraction effects in the masking approach to optical lithography with classical light limit the resolution of transferred images to the Rayleigh diffraction limit of $\lambda/4$, $\lambda$ being the optical wavelength. Boto *et al.* [3] showed that it is possible to breach this limit when maximally entangled light, say, for $n$ photons, interfere on the surface of a substrate capable of absorbing $N$ photons. A "proof of principle" experimental demonstration has recently been given [15]. In the present paper, we assume that our substrate absorbs only at some even photon number $N=2M$, $M=1,2,3\cdots$. We ignore the material issues of creating appropriate substrates, but see [3]. We restrict ourselves to the consideration of one dimensional lithography as in Ref. [3]. Our proposed lithographic procedure is illustrated schematically in Fig. 4. Here the relative phase shift $\phi$ between the two beams is $\phi=kx$ where $x$ is the lateral distance along the substrate and $k=2\pi/\lambda$ is the wave number. The state of the light beams just prior to the substrate is thus

$$|\psi\rangle=\frac{1}{\sqrt{2}}\sum_{m=0}^{\infty} C_{2m}[e^{2im\phi}|2m\rangle_a|0\rangle_b+e^{-i\Phi_{2m}}|0\rangle_a|2m\rangle_b]. \tag{21}$$

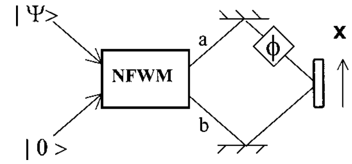


FIG. 4. Using the nonlinear four-wave mixer for photolithography. Here $\phi=2\pi x/\lambda$ where $x$ represents the lateral position across the substrate. The substrate is assumed to absorb an even number $N$ of photons.

On the surface of the substrate the field operator for the interfering beams is $e=a+b$. The deposition rate for $2M$ photon absorption is defined [3] as $\Delta_{2M,\gamma}=\langle\psi|\delta_{2M}|\psi\rangle$ where $\delta_{2M}=e^{\dagger 2M}e^{2M}/(2M)!$ is the dosing operator. Missing from this discussion, and also from that of Boto *et al.* [3], is any consideration of the cross sections for $2M$ photon absorption. We ignore them here as their likely effect is to decrease the overall deposition rate while our main interest is the breaching of the Rayleigh diffraction limit. Suppose for the moment that we have a state with all $C_{2m}=0$ except for $C_{2M}=1$. In this case, the deposition function is (assuming the light incident on the substrate at the grazing limit)

$$\Delta_{2M,\gamma}=[1+\cos(2M\phi+\Phi_{2M})]. \tag{22}$$

The spatial oscillation of this function indicates a resolution of $\Delta x=\lambda/8M$, a reduction by the photon number $2M$ below the Rayleigh limit $\lambda/4$. For the more general state of Eq. (20), again assuming a substrate absorbing only $2M$ photons, we obtain

$$\Delta_{2M,\gamma}=|C_{2M}|^2[1+\cos(2M\phi+\Phi_{2M})]+\sum_{m=M+1}^{\infty}|C_{2m}|^2\binom{2m}{2M}. \tag{23}$$

Note that states for which $m<M$ make no contribution and that those for which $m>M$ contribute to a constant term, the background.

This background term is a problem. It is independent of the phase $\phi$ and thus gives a uniform background exposure to the substrate. To illustrate how the presence of the background term effects this application of the squeezed and even coherent states, we show closed-form results that can be obtained from these states. In the case of two-photon absorption $(M=1)$, for the even coherent states we obtain

$$\Delta_{2,\gamma}=\frac{|z|^2}{2!}[1+\sin(2\phi)/\sqrt{\cosh(|z|)}]. \tag{24}$$

In fact, for these states it is easy to obtain the general result for a $2M$-photon absorbing substrate

$$\Delta_{2M,\gamma}=\frac{|z|^{2M}}{(2M)!}[1+\sin(2M\phi)/\sqrt{\cosh(|z|)}]. \tag{25}$$

For the case of the squeezed vacuum we obtain the two-photon deposition function

$$\Delta_{2,\gamma}=\frac{1}{2}\sinh^2 r\left[1+\frac{3}{2}\sinh^2 r-\frac{\sin(2\phi)}{\cosh^3 r}\right]. \tag{26}$$

(For $N$ photon absorption with $N=2M>2$, closed-form expressions for the deposition functions are more difficult to obtain in the case of the squeezed vacuum inputs than for the even coherent states and we do not pursue them.) We may characterize these results with the visibility $\mathcal{V}$ defined in the usual way so that for the even coherent state $\mathcal{V}_{\mathrm{ECS}}=1/\sqrt{\cosh(|z|)}$ and for the squeezed vacuum state $\mathcal{V}_{\mathrm{SV}}=1/\cosh^3 r$. Obviously for high-field strengths, the visibilities approach zero. Thus photolithography with either even coherent states or squeezed vacuum states will necessarily be restricted to weak beams.

The root of the problem, as mentioned above, is the presence of the background term in Eq. (23). As the field strength is increased, this term becomes very large, essentially washing out the effects of the first term. The effect of the background term may be minimized by resorting to weak fields but that in turn also decreases the overall deposition rate. To circumvent the problem one might prepare a truncated single-mode field state containing only a finite number of even photon states such that the $C_{2m}=0$ for $m>M$. In the case of an $N=2M$-photon-absorbing substrate the background term in Eq. (23) vanishes. Pegg, Phillips, and Barnett [16] have described a method for creating arbitrary traveling-wave optical states of the form $c_0|0\rangle+c_1|1\rangle$ by a procedure called the optical truncation of a state by projection synthesis (''quantum scissors''). Villas-Boas *et al.* [17] have extended the procedure to create states of the form $c_0|0\rangle+c_1|1\rangle+c_2|2\rangle+\cdots+c_N|N\rangle$. One could presumably truncate an even coherent state. Dakna *et al.* [18] have discussed a procedure for generating arbitrary states of a single-mode traveling-wave field. Thus by these procedures an appropriate quantum state containing only even number states could be created, which can subsequently be converted into superpositions of MES by the NFWM device. The techniques could even be used to generate even number states that may subsequently be converted into MES by a NFWM.

In conclusion, we have studied the generation of superpositions of maximally entangled two-mode photonic states by a nonlinear four-wave mixer and their application to interferometry and photolithography. Even numbered photon states are required for the NFWM device so we consider as inputs even coherent states and the squeezed vacuum states. We have found that for the purpose of interferometry, as inputs the even coherent states are superior to the squeezed vacuum states in that the former are useful over a wide range of phase differences whereas for the latter the phase uncertainty becomes degraded even for small values of the phase difference. However, it must be said that generating the even coherent states is a nontrivial task. But as pointed out in [4] and [5], a NFWM operating under a different condition than required to generate the MES, coupled with state reduction techniques, could be used to generate such states. A number of other techniques for generating the even coherent states have been discussed in the literature [12]. The general problem with generating the even coherent states is that some form of nonunitary process, such as state reduction, must be involved. Thus it will be difficult to produce such states continuously. The squeezed vacuum states on the other hand can easily be generated through the process of degenerate parametric down conversion. In low doses, required to minimize the background, these states may be useful for photolithography. We have shown that truncated states can be used to circumvent this difficulty.

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