We investigate the origin of the resolution improvement beyond the classical diffraction limit in quantum lithography. We show that the degree of the resolution improvement persists even when the pure entangled state of light initially prepared (the NOON state) is transformed to a mixed entangled state caused by phase and amplitude damping, regardless of the degree of entanglement remaining in the state. We also show that the resolution improvement beyond the classical diffraction limit can be achieved with a particular class of separable states of light. The resolution improvement in this case can be traced to entanglement (of the NOON type) that may be considered to be embedded in the separable state. We conclude that it is the existence of entanglement (of the NOON type) in the state of light used, not details of entanglement such as the degree of entanglement and relative weight of entanglement in the state, that determines the degree of the resolution improvement in quantum lithography. The visibility of the pattern and the total deposition rate, however, depend on the details of entanglement.

KEYWORDS: quantum lithography, NOON state, resolution improvement

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The Role of Entanglement in Quantum Lithography

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1. Introduction

Optical lithography refers to a process of projecting the image of patterns on substrates using visible or ultraviolet light. Due to diffraction of light being used, the highest resolution one can achieve in the standard classical lithography is $\lambda/2$ ($\lambda$ being the optical wavelength), the limit set by the Rayleigh criterion. Recently, much attention has been given to the technique of quantum lithography which is capable of beating the classical diffraction limit. As first proposed by Boto et al. in 2000, the idea is to use nonclassical states of photons entangled between the vacuum and N-photon states ("NOON" states), which enables one to write features of minimum size $\lambda/2N$ on a N-photon absorbing substrate. A proof-of-principle experimental demonstration of quantum lithography has been performed for the case $N = 2$ by D’Angelo et al. in 2001. The original quantum lithographic scheme of Boto et al., however, is not without difficulties, one of which is the difficulty of generating the NOON state, which, for the case $N = 2$, can be provided by combining two photons generated by spontaneous parametric down conversion at a 50/50 beam splitter. It has been suggested that a high-gain parametric amplifier can replace a parametric converter, because the output of a high-gain parametric amplifier still possesses the quantum property associated with two- and multi-photon interference needed for the resolution enhancement. This idea has recently been verified experimentally, which opens a way to an easier and wider implementation of the quantum lithographic scheme. To a certain extent, the difficulty of generating the NOON state can be bypassed by utilizing a time-reversal technique. For example, phase super-resolution with entangled photons has been demonstrated based on probabilistic detection, rather than generation, of the NOON state. As this technique utilizes time reversal, however, it cannot be directly applied to quantum lithography which requires the presence of the NOON state.

The resolution enhancement in quantum lithography can be understood in terms of de Broglie wavelength of entangled photons. The de Broglie wavelength of an entangled photon pair has been measured and shown to lead to sub-Rayleigh interference patterns.

The use of quantum entangled states of photons is not an absolute requirement to go beyond the Rayleigh diffraction limit in lithography. There exist several proposals and experimental demonstrations for methods for increasing the resolution of an optical lithographic system employing only classical light. Already in 1994, Ooki et al. proposed to use nonlinear absorbing materials and a multiple exposure technique to obtain resolution enhancement. In 1999, Yablonovitch and Vrijen proposed an interference technique with multiple frequency beams to achieve doubling of the spatial resolution, which was experimentally demonstrated in 2002. In 2004, Bentley and Boyd achieved experimentally 2- and 3-fold enhancement of resolution with classical light by using a nonlinear optical interferometric method exploiting, as suggested by Ooki et al., the nonlinear response of the recording medium. The method is easier to implement than the method of Yablonovitch and Vrijen. Further experimental demonstrations of this method were reported by Chang et al. and by Khoury et al. Similar methods based on classical pulses and classical thermal light were also proposed. Very recently, a different scheme using classical light was proposed, in which sub-Rayleigh fringes are obtained by correlating wave number and frequency in a narrow band multiphoton detection process, as the frequency-selective measurement process allows one to simulate quantum entanglement. More recently, a method using correlations between incoherent photons, which relies on neither photon entangled states nor multiphoton absorption, was proposed.

The resolution enhancement in quantum lithography is believed to originate from the peculiar quantum nature of entangled states. On the other hand, the techniques employing classical light rely primarily on the nonlinear response of the recording medium and thus appear largely unrelated to
quantum features of light. It should, however, be noted that nonlinear absorption in these techniques can be viewed to play the role of extracting from classical light used the quantum property necessary for the resolution enhancement so that the unusual quantum statistics of entangled photons is mimicked. The question then arises exactly what properties of light are required to beat the Rayleigh diffraction limit. One way to attack this question is to look at the problem from the perspective of quantum lithography and ask what the exact role of entanglement is in quantum lithography. Are subwavelength fringes in quantum lithography obtained only with pure entangled states of photons? Can the Rayleigh diffraction limit be overcome with mixed entangled states of photons or even with separable states of photons?

In this paper, we provide answers to the above questions and thereby clarify the exact role of entanglement in quantum lithography. We first calculate the interference pattern when mixed entangled states of photons are used. This is a practically important issue, because a pure entangled state, e.g., the NOON state, in reality goes inevitably to a mixed entangled state due to unavoidable interaction with environments. We22,23 show that, rather surprisingly, the peak-to-peak separation of the interference pattern when mixed entangled states of photons are used.

In §4 we introduce a separable state of light which is capable of producing interference patterns that yield the same degree of resolution enhancement obtained with the pure NOON state. We then introduce a particular type of separable states of photons which yields the same degree of resolution enhancement as the pure quantum entangled states. We show that the resolution improvement in this case results from entanglement which may be considered to be “embedded” in the separable state.

In §2 we give a brief review of quantum lithography using the NOON state, a pure entangled state of light. We then study in §3 the problem of how the interference pattern changes when the NOON state (with \( N = 2 \)) decays to a mixed entangled state due to phase damping and amplitude damping arising from interaction with the environments. In §4 we introduce a separable state of light which is capable of producing interference patterns that yield the same degree of resolution enhancement obtained with the pure NOON state. Finally §5 presents the conclusion of our work.

2. Quantum Lithography with Pure Entangled States of Light

The interferometric lithography setup we consider is illustrated in Fig. 1. It is the same system that Boto et al.24 considered. One photon each is sent to the symmetric, lossless, 50/50 beam splitter through each of its two input ports A and B. A pair of path-entangled photons are then produced in the output ports A’ and B’. A phase shifter, the action of which is described by a unitary operator \( e^{-i\varphi a^\dagger a} \), is placed in the path A’. The phase shift \( \varphi \) models the phase difference between two counterpropagating beams at different positions \( x \) of the substrate through the relation \( \varphi = 2kx \).

The state of the photons in modes C and D that interfere on the substrate is given by

\[
|\psi\rangle_{CD} = \frac{1}{\sqrt{2}} (e^{-i2\varphi}|2\rangle_c|0\rangle_D + |0\rangle_c|2\rangle_D). \quad (1)
\]

The two-photon absorption rate at the surface defined as

\[
\Delta_2 = cD(\langle \hat{\delta}_2 | \hat{\delta}_2 \rangle |_{CD}.
\]

where \( \delta_2 = (\hat{c}^\dagger \hat{c}^\dagger + \hat{d}^\dagger \hat{d}^\dagger) / \sqrt{2} \) and \( \hat{c} \) and \( \hat{d} \) are annihilation operators for a photon of mode C and D, respectively, is then easily calculated to be

\[
\Delta_2 = 1 + \cos 2\varphi. \quad (3)
\]

Equation (3) indicates that resolution is enhanced by a factor of 2. A straightforward generalization to the \( N \)-photon entangled “NOON” state

\[
|\psi\rangle_{CD} = \frac{1}{\sqrt{2}} (e^{-iN\varphi}|N\rangle_c|0\rangle_D + |0\rangle_c|N\rangle_D) \quad (4)
\]

gives an \( N \)-fold enhancement of resolution.

3. Quantum Lithography with Decohered Mixed Entangled States of Light

Although in the previous section, we have assumed a pure path-entangled state for the photons that enter the surface of the substrate, in reality interaction with the environments is unavoidable, causing the photons to be in a mixed state. We study the effects of decoherence arising from interaction with the environments upon the interference pattern recorded on the substrate.

Let us first consider phase damping, which is described by the state transformation25

\[
|0\rangle_S|x_0\rangle_E \rightarrow \sqrt{1 - p} |0\rangle_S|x_0\rangle_E + \sqrt{p} |0\rangle_S|x_2\rangle_E, \quad (5a)
\]

\[
|2\rangle_S|x_0\rangle_E \rightarrow \sqrt{1 - p} |2\rangle_S|x_0\rangle_E + \sqrt{p} |2\rangle_S|x_1\rangle_E, \quad (5b)
\]

where the subscripts \( S \) and \( E \) refer, respectively, to the system (mode A’ or B’ for the setup of Fig. 1) and the environment, and \( p \) is the probability for the environment to be transformed from its initial state \( |x_0\rangle_E \) into the state \( |x_2\rangle_E \) if the system \( S \) is in the state \( |0\rangle_S \) and into the state \( |x_1\rangle_E \) if the system \( S \) is in the state \( |2\rangle_S \). With phase damping taking
The logarithmic negativity of this state is given by
\[
\rho_{\text{CD}} = \frac{1}{2} \left[ |02\rangle \langle 02| + (1 - p)^2 e^{2i\phi} |02\rangle \langle 02| + (1 - p)^2 e^{-2i\phi} |02\rangle \langle 20| + |20\rangle \langle 02| \right]_{\text{CD}}.
\]
Note that phase damping affects only the off-diagonal elements of the density matrix.

The degree of entanglement associated with the mixed state (6) can be estimated by the logarithmic negativity\(^{26-28}\) defined as
\[
\mu = \log_2 \left( \sum_i |\lambda_i| \right),
\]
where \(\lambda_i\)'s are the eigenvalues of the matrix \(\rho_{\text{CD}}^\text{Tc}\) (or \(\rho_{\text{CD}}^\text{Td}\)) obtained by partial transposition of \(\rho_{\text{CD}}\) with respect to the system C (or the system D).\(^{26,27,29}\) A positive value of \(\mu\) implies that the state is entangled. A straightforward calculation yields, for the state (6),
\[
\mu = \log_2 (1 - p^2) + 1.
\]
As phase damping progresses, \(\mu\) decreases monotonously from its initial maximum value of 1 toward 0. At \(p = 1\), \(\mu\) becomes 0 and entanglement disappears.

The two-photon absorption rate \(\Delta_2\) with phase damping taken into account can easily be calculated. By substituting eq. (6) into \(\Delta_2 = \text{T}_{\text{CD}}(\Delta_2_{\text{CD}})\), we obtain
\[
\Delta_2 = 1 + (1 - p)^2 \cos 2\phi.
\]
Equation (9) indicates that the resolution enhancement by a factor of 2 persists even in the presence of phase damping. On the other hand, the visibility \(V\) of the interference pattern is given by
\[
V = \frac{(\Delta_2)_{\text{max}} - (\Delta_2)_{\text{min}}}{(\Delta_2)_{\text{max}} + (\Delta_2)_{\text{min}}} = (1 - p)^2
\]
and thus decreases as phase damping progresses.

We next consider amplitude damping, which is described by the state transformation\(^{25}\)
\[
|0\rangle_1 |0\rangle_2 \rightarrow |0\rangle_1 |0\rangle_2 E, \quad (10a)
\]
\[
|1\rangle_1 |0\rangle_2 \rightarrow \sqrt{1-p} |1\rangle_1 |0\rangle_2 + \sqrt{p} |0\rangle_1 |1\rangle_2 E, \quad (10b)
\]
\[
|2\rangle_1 |0\rangle_2 \rightarrow (1-p) |2\rangle_1 |0\rangle_2 + \sqrt{2p} (1-p) |1\rangle_1 |1\rangle_2 E + p |0\rangle_1 |2\rangle_2 E, \quad (10c)
\]
where \(p\) now represents the probability of losing a photon. The density matrix for the state of the photons that interfere on the substrate in the presence of amplitude damping can be obtained through a straightforward calculation as
\[
\rho_{\text{CD}} = p^2 |00\rangle \langle 00| - p (p - 1) (|01\rangle \langle 01| + |10\rangle \langle 10|)
+ \frac{1}{2} (p - 1)^2 |02\rangle \langle 02| + e^{2i\phi} |02\rangle \langle 20| + e^{-2i\phi} |20\rangle \langle 02| + |20\rangle \langle 20|).
\]
The logarithmic negativity of this state is given by
\[
\mu = \log_2 \left( \frac{1}{2} p^2 + \frac{1}{2} \sqrt{2} p^3 + 1 - 4p + 6p^2 - 4p^3 \right)
+ \frac{1}{2} p^2 - \frac{1}{2} \sqrt{2} p^3 + 1 - 4p + 6p^2 - 4p^3
\]
and decreases monotonously from \(\mu = 1\) at \(p = 0\) to \(\mu = 0\) at \(p = 1\). We see that entanglement exists as long as \(p < 1\) and it disappears at \(p = 1\).

The two-photon absorption rate \(\Delta_2\) with amplitude damping taken into account can be obtained by a straightforward calculation:
\[
\Delta_2 = (p - 1)^2 (1 + \cos 2\phi).
\]
Here, we see that, as amplitude damping progresses and the probability \(p\) increases from 0 to 1, doubling of the resolution and the unit visibility remain unchanged. Thus, amplitude damping affects neither of the two. The total rate of absorption, however, decreases toward 0 as \(p\) increases to 1.

4. Quantum Lithography with Separable States of Light

It is shown in the previous section that the degree of resolution enhancement is preserved even if entanglement is degraded due to phase or amplitude damping. In order to shed more light on the question of the exact role of entanglement in quantum lithography, we introduce in this section a separable state of light which yields the same degree of resolution enhancement as the NOON state and discuss where the resolution enhancement originates from in this case of a separable state.

Let us consider the situation in which each of the modes A' and B' of Fig. 1 contains a linear superposition of the vacuum and two-photon states, (1/\(\sqrt{2}\))(|0\rangle + |2\rangle). The state evolves after passage through the phase shifter into
\[
|\psi\rangle_{\text{CD}} = \frac{1}{\sqrt{2}} (|0\rangle_0 c + e^{-2i\phi} |2\rangle_0 c) \otimes \frac{1}{\sqrt{2}} (|0\rangle_D + |2\rangle_D)
= \frac{1}{2} |00\rangle c_0 d + \frac{1}{2} e^{-2i\phi} |22\rangle c_0 d
+ \frac{1}{2} (|00\rangle c_2 d + e^{-2i\phi} |2c_0\rangle c_2 d). \quad (14)
\]
The two-photon absorption rate at the surface sensitive to two-photon absorption can easily be calculated to be
\[
\Delta_2 = 3 + \frac{1}{2} \cos 2\phi. \quad (15)
\]
Equation (15) indicates that, even with the separable state of eq. (14), doubling of the resolution is achieved, although the visibility of the interference pattern drops to 1/6.

It is not difficult to see where the enhanced resolution and the low visibility originate from. When two photons are absorbed at the surface of the substrate, the state |\psi\rangle_{\text{CD}} of eq. (14) is transformed to the state
\[
|\psi\rangle_{\text{CD}} = \frac{1}{\sqrt{2}} e^{2i\phi} |\psi\rangle_{\text{CD}}
= \frac{1}{2} (1 + e^{-2i\phi}) |00\rangle
+ \frac{1}{2} e^{-2i\phi} (|22\rangle + 2\sqrt{2} |11\rangle + |20\rangle). \quad (16)
\]
Note that it is the first term on the right-hand-side of eq. (16) that yields the interference term responsible for the resolution enhancement. This term originates from the "entangled part"
of the state (14), i.e., from the term \( (1/2) |0\rangle_C |2\rangle_D + e^{-i2\varphi} |2\rangle_C |0\rangle_D \). One can thus say that entanglement embedded in the separable state of eq. (14) is the source for the resolution enhancement. The second (last) term on the right-hand side of eq. (16), on the other hand, is responsible for the low value of the visibility. It is actually the relative weight of this second term with respect to the first term that determines the visibility. Considering that this second term originates from the term \( (1/2) e^{-i2\varphi} |2\rangle_C |0\rangle_D \) of eq. (14), one can easily see that a higher visibility can be obtained by choosing the initial state that has a lower weight of the two-photon state. To take a simple example, let us consider the case where \( |\psi\rangle_{CD} \) is given by

\[
|\psi\rangle_{CD} = (a|0\rangle_C + be^{-i2\varphi}|2\rangle_C) \otimes |a|1\rangle_D + b|2\rangle_D.
\]  

where, for simplicity, we assume \( a \) and \( b \) are real \((a^2 + b^2 = 1)\). The two photon absorption rate in this case is given by

\[
\Delta_2 = 2a^2b^2 + 10b^4 + 2a^2b^2 \cos 2\varphi.
\]  

Clearly, the resolution is independent of \( a \) and \( b \), but the visibility is not, as it is given by \( V = a^2/(5 - 4a^2) \). Thus, a high visibility can be obtained without sacrificing resolution simply by taking the coefficient \( a \) to be close to 1, thus by choosing the entangled part of the state (14) to have a relatively high weight compared to the two-photon part \( |2\rangle_C |2\rangle_D \). As the coefficient \( a \) is brought closer to 1, however, the total rate of two-photon absorption decreases toward 0, because the state \( |\psi\rangle_{CD} \) approaches the vacuum state \( |0\rangle_C |0\rangle_D \).

The treatment given here can easily be generalized to the case of a separable state with each of the two modes C and D in a superposition of the vacuum and N-photon states. For \( |\psi\rangle_{CD} \) given by

\[
|\psi\rangle_{CD} = \frac{1}{\sqrt{2}} (|0\rangle_C + e^{-iN\varphi}|N\rangle_D) \otimes \frac{1}{\sqrt{2}} (|0\rangle_D + |N\rangle_D).
\]  

The absorption rate on the substrate sensitive to N-photon absorption can be shown to be given by

\[
\Delta_N = \frac{1}{4} + \frac{N}{4} \sum_{k=0}^{N} (\chi_{C_k})^3 + \frac{1}{2} \cos N\varphi.
\]  

where \( \chi_{C_k} = \sqrt{N}/k! (N - k)! \). Thus, the resolution is enhanced by \( N \) times, but the visibility decreases as \( N \) is increased.

5. Conclusion

The purpose of this work is to investigate the role of entanglement in quantum lithography. We have demonstrated that mixed damped NOON states of photons are capable of producing patterns with N-fold enhancement of resolution just like the pure NOON states. Furthermore, the N-fold enhancement of resolution can be achieved even with a particular class of separable states in which each of the two photon modes contains a superposition of the vacuum and N-photon states. The resolution enhancement in this case originates from entanglement embedded in the separable state. Considering that the same N-fold enhancement of resolution is obtained with mixed damped NOON states and with the separable states containing entanglement embedded in them as well as with the pure NOON states, we can conclude that the resolution improvement beyond the Rayleigh diffraction limit can be attributed just to the existence of entanglement in the state of light used. Depending on details of entanglement such as the degree of entanglement and the relative weight of entanglement, the visibility of the interference pattern and the total rate of absorption may vary, but resolution is independent of such details. As long as the state of light used contains the NOON state, N-fold enhancement in resolution is obtained, regardless of whether the NOON state contained is pure, mixed, or even embedded in a separable state.

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1) In this paper, the term “resolution” is used to indicate the fringe interval which determines the minimal resolvable feature size.