Entangled-State Lithography: Tailoring any Pattern with a Single State

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We demonstrate a systematic approach to Heisenberg-limited lithographic image formation using four-mode reciprocal binomial states. By controlling the exposure pattern with a simple bank of birefringent plates, any pixel pattern on a \((N+1) \times (N+1)\) grid, occupying a square with the side half a wavelength long, can be generated from a \(2N\)-photon state.

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When two coherent plane waves of wavelength \(\lambda\) are made to overlap, a striped intensity distribution with perfect sinusoidal modulation is formed. A simple application of the Rayleigh criterion shows then that the minimum resolvable feature size occurs at a spacing of \(\lambda/2\), which is the best resolution that can be achieved classically and is usually known as the diffraction limit.

On the contrary, quantum fields allow the modulation period to be as small as \(\lambda/(2N)\), where \(N\) is the average number of photons of the two interfering modes. The origin of the sub-diffraction resolution can be envisioned as the photons clustering into quasi-particles with a de Broglie wavelength inversely proportional to the (relativistic) mass of the \(N\)-photon quasi-particle [1]. This is the basis of the so-called Heisenberg-limited interferometry, that has been experimentally demonstrated [2, 3, 4]. In addition, the modulation pattern of two interfering quantum fields is, in general, not sinusoidal, but can take many shapes, a fact we shall exploit later.

In this respect, image formation can be seen as a multimode generalization of interferometry. Nowadays, it is recognized that for a variety of imaging techniques we can have a feature resolution that is ultimately limited not by diffraction, but by the quantum fluctuations of the light beams used in the experiments [5].

In all cases, the existence of the standard diffraction limit can be traced back to the fact that light is treated as a classical beam or, which is equivalent, as a stream of uncorrelated photons. It has been known for some time that entangled photon pairs show unusual resolving facts, but only very recently it has been proposed the use of entanglement to increase the resolution indefinitely, a fact that opened the possibility of performing quantum optical lithography [6].

In this Letter we go one step further in this promising field and discuss the use of reciprocal binomial states instead of entangled number states. At first sight, this could be seen as a curiosity, but one must take into account that these states are the basis of the projection synthesis method [7] since they are the reference to be used in a beam splitter such that the photon counts at the two outputs lead to the experimental determination of the expectation value of any projector formed by the first \(N+1\) number states. Therefore, they offer the capability of tailoring any desired pattern, as we shall show below.

A great advantage of our method is that only one entangled state needs to be generated. All other necessary states can be produced from the first by means of a small bank of phase plates with a prescribed birefringence. Moreover, it also works for any number of photons \(N\) and offers a very intuitive and simple means of determining the exposure sequence to generate any pixelated pattern on a \((N+1) \times (N+1)\) grid, occupying a square with a half wavelength long side.

Our goal is to establish how we can create arbitrary 2D patterns on a suitable substrate. Suppose we have two counter-propagating beams 1 and 2, in some direction we shall denote as the axis \(X\), that propagate at grazing incidence over a substrate of side \(\lambda/2\) coated with lithographic resist (in the following we will refer to the resist as the film). In general, we have to take into account the mode shapes, but when restricted to a length \(\lambda\) this problem does not arise, provided the coherence lengths of the wavepackets are much longer than the side of the film. In reality, this is a very lax requirement since it means that a wavepacket with a coherence length of, say, 100\(\lambda\) meets this criterion by far. In the orthogonal \(Y\) direction we have two other beams 3 and 4, satisfying the same conditions. The square film is situated in the region where these four beams overlap.

If we restrict ourselves to consider \(N\)-photon states in the four modes, a quick combinatorial calculation gives that there are \((N+1)(N+2)(N+3)/3!\) such pure states. In principle, it would be possible to generate this number of distinct patterns (but due to the setup’s rotational symmetry, degeneracies would occur). However, different nonlinear Hamiltonians would be required to generate all the different patterns. In Ref. [8] the lithographic film absorption is modeled by an \(N\)-photon absorption process. In this way, higher-order interference effects are naturally brought out. Unfortunately, the \(N\)-photon absorption...
cross section decreases very rapidly with increasing $N$ in all materials. However, to harness the de Broglie wavelength of $N$-photon wavepackets all the photons in the wavepacket must collectively interfere and, therefore, $N$-photon absorption processes seems be needed. In addition, for the quantum lithography process to make sense, the photosensitive “grains” in the film must be much smaller that the shortest de Broglie wavelength encountered in the exposure process. Therefore, from the point of view of a “grain”, the photon packets in the respective modes will be indistinguishable in spite of their different linear momenta. Consequently, the Hamiltonian governing the absorption can be written as

$$\hat{H} \propto (\hat{a}_1^\dagger + \hat{a}_1 + \hat{a}_2^\dagger + \hat{a}_2)^N(\hat{a}_1 + \hat{a}_2 + \hat{a}_3 + \hat{a}_4)^N,$$  

(1)

where $\hat{a}_i$ is the annihilation operator of mode $i$.

For the moment, let us restrict the problem to one dimension along the axis $X$. According to Ref. [3], the deposition rate in the substrate $\Delta_N$ is then given by the expectation value of the operator $e^{i\lambda x}e^N/N!$, where $\lambda = (a_1 + a_2)/\sqrt{2}$. But, this is the action of a 50:50 beam splitter for modes 1 and 2, and inspired by the projection synthesis method, it is natural to use two-mode $N$-photon reciprocal binomial states [7]

$$|\psi^{(N)}\rangle = \frac{1}{\sqrt{N}} \sum_{n=0}^{N} \sqrt{n!(N-n)!} |n, N-n\rangle,$$  

(2)

where $|n, N-n\rangle = |n\rangle_1 \otimes |N-n\rangle_2$ and $N = \sum_{n=0}^{N} n!(N-n)!$ is a normalization factor.

Since modes 1 and 2 impinge over the film in antiparallel directions, the accumulated phase of mode 1 at a distance $x/\lambda$ from the left edge of the film will be

$$\hat{U} = \exp(ik\hat{n}_1 \lambda x) = \exp(i2\pi x \hat{n}_1),$$  

(3)

where $k = 2\pi/\lambda$ and $\hat{n}_1 = \hat{a}_1^\dagger \hat{a}_1$, while mode 2 will have accumulated the phase

$$\hat{U} = \exp[ik\hat{n}_2 \lambda(1-x)] = \exp[i2\pi(1-x) \hat{n}_2]$$  

(4)

at the same location. Using these free-space unitary propagation operators, we find that at the location $x$, the state [3] is transformed into

$$|\psi^{(N)}_x\rangle = \frac{1}{\sqrt{N}} \sum_{n=0}^{N} e^{i2\pi x(2n-N)} \sqrt{n!(N-n)!} |n, N-n\rangle.$$  

(5)

Calculating the pattern deposition rate $\Delta_N$ for the state $|\psi^{(N)}_x\rangle$, we find that

$$\Delta_N \propto \left| \sum_{n=0}^{N} e^{i2\pi x(2n-N)} \right|^2 \propto \sin^2\left\{2(N+1)x - \frac{\ell}{N+1}\right\}/(N+1)^2 \sin^2\left\{2\pi x + \frac{\ell}{N+1}\right\}.$$  

(6)

We observe that $\Delta_N$ has a highest oscillation period in $x$ of $1/[2(N+1)]$ and an overall periodicity of $1/2$, corresponding to the physical lengths $\lambda/[2(N+1)]$ and $\lambda/2$, respectively. The former is the shortest oscillation period possible using $N$-photon states [8].

Now assume that we translate the substrate a distance $\lambda/[4(N+1)]$ to the left, and that we phase-shift mode 1 by $2\pi\ell/(N+1)$ ($\ell = 1, 2, \ldots, N+1$) relative to mode 2. We denote the ensuing state $|\psi^{(N,\ell)}_x\rangle$. A simple way of doing this would be to let mode 1 and 2 be spatially and temporally degenerate modes, but with orthogonal polarizations, see Fig. [1]. Insertion of phase plates with a birefringence corresponding to an optical path difference of $\lambda/((N+1)$ and with their principal axes parallel to the polarization directions of the modes, would provide the needed relative phase shift. The deposition rate after these additional modifications are taken into account can readily be calculated to be

$$\Delta_N \propto \sin^2\left\{2(N+1)x - \ell + 1/2\right\}/(N+1)^2 \sin^2\left\{(2x - \ell + 1/2)\pi\right\}.$$  

(7)

Let us now examine the properties of the deposition rate function. The function has one peak, roughly $1/[2(N+1)]$ wide in units of $\lambda$, and for each successive state $|\psi^{(N,\ell)}_x\rangle$, where $\ell = 1, 2, \ldots, N+1$, the peak is displaced a distance $1/[2(N+1)]$ to the right. Hence, if we “pixelate” the substrate along the $X$ direction into $N+1$ pixels, each state will expose (or deposit) one pixel, with a negligible deposition rate outside the pixel. Note, in particular, that the deposition rate due to state $|\psi^{(N,\ell)}_x\rangle$ at the center of pixel $\ell_i$ is identically zero for all states with $\ell \neq \ell_i$. That is, the deposition penalty at the center of each pixel is identically zero, independent of which other pixels are exposed.

Assume now that we have a similar exposure apparatus along the $Y$ direction, i.e., a second synchronized generator of the state [3] in modes 3 and 4, followed by a bank of birefringent plates, a polarizing beam splitter, and rigid mirrors. The state at the point $(x, y)$ of the substrate would then be in a $2N$-photon four-mode product state $|\psi^{(N,\ell_x)}_x \otimes \psi^{(N,\ell_y)}_y\rangle$ of the substrate along the $X$ direction into $N+1$ pixels, each state will expose (or deposit) one pixel, with a negligible deposition rate outside the pixel. Note, in particular, that the deposition rate due to state $|\psi^{(N,\ell)}_x\rangle$ at the center of pixel $\ell_i$ is identically zero for all states with $\ell \neq \ell_i$. That is, the deposition penalty at the center of each pixel is identically zero, independent of which other pixels are exposed.

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The price we pay for preparing this $2N$-photon product state, rather than a more general state, is that we can only deposit $(N+1)(N+1)$ different patterns, instead of close to $(2N+1)(2N+2)(2N+3)/3!$. However, the patterns are essentially mutually exclusive, so by considering mixed two-mode reciprocal binomial states we can build any one of the $2(2N+1)^2$ possible different patterns.

In Fig. 2 we have simulated the deposition of a serpentine pattern exposed across the sample. The pixels $(2,1)$, $(2,2)$, $(2,3)$, $(2,4)$, $(3,4)$, $(4,4)$, $(5,4)$, $(5,5)$, $(5,6)$, and $(5,7)$ have been exposed by preparing a superposition of the states with corresponding $\ell_x$ and $\ell_y$. Additionally, we have exposed the pixel $(6,4)$ to demonstrate that nonexposed pixels sitting between two exposed ones, receive a negligible deposition penalty (identically zero at the pixel center). In practice, the simplest way to do this is to expose the substrate sequentially and for equal times, first by one of the states, then by the other. The only experimental adjustment that needs to be done between the exposures is a rearrangement of the bank of birefringent plates. The bank need not be large: to accomplish any of the needed $N$ relative phase-differences we need only $\log_2 N$ (or actually the smallest integer greater or equal to $\log_2 N$) birefringent plates. That is, for a $1025 \times 1025$ pixel pattern we need only 10 birefringent plates in each bank. The inset shows the exposure along the central ridge. We see that the exposure along the ridge has ripples, but the minimum is still 89% of the maximum exposure. We also see that the exposure penalty elsewhere is small (smaller than 12% of the maximum exposure). Taking the nonlinearity, with respect of the exposure dose, of the optical density of a lithographic film after development, such a ripple is acceptable. We also see one undesirable consequence of the $\lambda/2$ periodicity of the deposition rate. The pixel $(5,7)$ will partly expose the pixel $(5,1)$. This problem is fundamental but can be overcome by sacrificing some of the film area and considering only the $(N-1)$ central points.

In Fig. 3 we have simulated the deposition of a pattern going diagonally across the sample. This is the worst case using the proposed pixelation method. The inset shows the deposition rate along the center of the diagonal ridge. As expected, the deposition rate ripple is large, the minima are only 35% of the maxima. This is unacceptable. To correct this flaw, we shift the pattern along both axes by half a pixel. This can be accomplished either by translating the substrate by $\lambda/[4(N+1)]$ in both the $X$ and $Y$ directions, or by inserting a birefringent $\lambda/[2(N+1)]$-plate at both banks. This allows us to place “intermediate” pixels along the diagonal, filling in the deposition rate minima. In Fig. 4 we have simulated the same pattern by exposing pixels $(2,1)$, $(2,2)$, and $(5,7)$ with a full exposure dose, pixels $(2,3)$ and $(5,6)$ with a relative exposure dose of 0.83, pixels $(3,4)$ and $(4,5)$ with a relative exposure dose of 0.66, and added intermediate pixels at pixel positions $(2,5,3,5)$, $(3,5,4,5)$, and $(4,5,5,5)$, at a relative exposure dose of 0.66. The figure inset shows the deposition rate along the diagonal ridge, demonstrating that very good patterns can be formed in this way. The ridge’s deposition rate maximum is 1.04, and the minimum is 0.90 of the deposition rate of an isolated pixel exposed with the nominal dose.

It is worth discussing what the experimental challenges with quantum lithography are. We see three main hurdles. The first is to develop a reasonably sensitive $N$-photon absorption film. Such a film seems to be the only way to fully capitalize on the de Broglie wavelength of photon packets, and our proposal will share this difficulty with any other proposal. The second hurdle is how to generate the reciprocal binomial states. Today, no generator of such a state with $N > 2$ exists, while for $N = 2$ a two-photon source and an appropriate beam splitter can be used. However, at least one proposal to do this job, using atom-photon interaction, is at hand. In, e.g., the proposal of Boto et al. [5], each pattern needs a different, and typically equally complicated, state. Unfortunately, to make a generator of an infinite (or at least large) set of different entangled states seems impossible with present technology. Thirdly, the exposed area will be very small in comparison to the wafers, and even the chips, presently manufactured in the semiconductor industry. However, using only two modes along each direction will always produce periodic patterns. To increase the spatial repetition period, while keeping the resolution, more modes are needed. This is not impossible, but at least in our opinion, one must first establish a procedure for how to generate arbitrary patterns in a systematic manner. We hope this paper can serve as a basis for this line of development.

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FIG. 1: A schematic showing the proposed setup to expose the film in the $X$ direction. PBS denotes a polarization beam splitter. It is assumed that a synchronous state generator, a second birefringent phase-plate bank, and appropriate mirrors are also arranged in the $Y$ direction.

FIG. 2: Calculated deposition rate for a serpentine pattern made from a two six-photon two-mode reciprocal binomial states. We have also deposited in the pixel (6, 4). The inset shows the deposition profile along the line crossing the center of pixels (5, 1) and (5, 7).

FIG. 3: Calculated deposition rate for a S-curve made from two six-photon two-mode relative phase states. The inset shows the deposition profile along the diagonal, hilly ridge. The profile goes from pixel (1, 2) to pixel (6, 7).
FIG. 4: An improved deposition rate function, where intermediate pixels have been placed to make the diagonal ridge smoother. The inset shows the deposition along the ridge. The result is a substantial improvement from the previous figure.
\[ |\Psi_x^{(N)} \rangle \]
Generator

Bank of phase plates

\( \lambda/2 \) plate at 45°

PBS

Photographic film

\( Y \)
\( X \)
