Nonlinear optical lithography with ultra-high sub-Rayleigh resolution

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Abstract: A nonlinear optical, interferometric method for improving the resolution of a lithographic system by an arbitrarily large factor with high visibility is described. The technique is implemented experimentally for both two-fold and three-fold enhancement of the resolution with respect to the traditional Rayleigh limit. In these experiments, an N-photon-absorption recording medium is simulated by N^{th} harmonic generation followed by a CCD camera. This technique does not exploit quantum features of light; this fact suggests that the improved resolution achieved through use of "quantum lithography" results primarily from the nonlinear response of the recording medium and not from quantum features of the light field.

OCIS codes: (190.4180) Multiphoton processes; (110.5220) Photolithography.

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The ultimate limit to the resolution of any optical imaging or lithographic system is set by diffraction and is often described in terms of the Rayleigh criterion [1]. For both traditional incoherent mask projection lithography and interferometric lithography [2], the ideal limit to the feature size (peak-to-peak) that can be recorded is $\lambda/2$. Recently, much interest has been given to the proposal to use quantum-entangled photons in conjunction with a recording medium that functions by means of *N*-photon absorption to write lithographic features with a resolution *N* times better than the Rayleigh limit [3-6]. One proof-of-principle experiment has been reported [7] for the case N=2 in which the properties of the 2-photon recording medium were mimicked by an electronic coincidence circuit. This increased resolution can be understood from the point of view that de Broglie wavelength of an entangled photon pair is half the conventional wavelength of either photon [8, 9]. However, the practical implementation of quantum lithography using currently proposed methods is extremely challenging, in large part because of the conflicting requirements that the optical fields falling onto the recording medium be sufficiently strong to induce multiphoton absorption yet be so weak as to show strong quantum features. In the present Letter, we describe a procedure that can also increase the resolution by a factor of *N* but relies only on the classical properties of laser light and thus can be implemented using intense laser beams. This method thus lends itself for use in practical photolithographic systems.

It is important to realize that the resolution enhancement achievable by any of the currently proposed techniques, whether classical or quantum, results solely from the properties of the *N*-photon absorption process. When a spatially modulated light field falls onto such an absorber, the resulting excitation pattern consists of all spatial frequency components ranging from the fundamental spatial frequency to *N*-times this frequency. The key to resolution enhancement is thus to eliminate the low spatial frequency components while retaining the high frequency components.

In the proposal of Boto et al. [3], the nature of the quantum interference between two entangled photons ensures that none of the low spatial frequency components is recorded. However, it does this only when the optical field strength is so weak that at a given time only *N* total photons are incident on the *N*-photon absorber. This condition imposes a strong limitation on deposition rate. If one increases the strength of the optical field, the visibility of the recorded pattern becomes greatly reduced. The origin of this degraded contrast is the presence of unwanted spatial harmonic components in the deposition rate [10, 11]. Because of the reduced fringe visibility at high deposition rates, the general usefulness of that method is quite limited.

The method presented in this Letter can be implemented straightforwardly and leads in principle to an arbitrarily large improvement in resolution. In this Letter we present laboratory results that demonstrate resolution improvements of up to three times the Rayleigh limit. Since this method relies only on classical aspects of the light field, the field strength can be arbitrarily large, thus allowing for very high deposition rates.

The technique is shown schematically in Fig. 1. An intense laser pulse is divided into two equal components at a beamsplitter. One component is shifted in phase with respect to the other, and the two components are then allowed to interfere on a lithographic plate that functions by means of *N*-photon absorption. To achieve enhanced resolution by a factor of M, where $M \leq N$, this is process is repeated M times, with the

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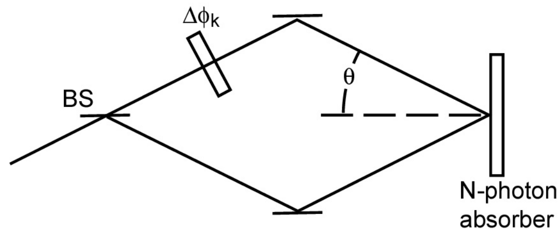

Fig. 1. Schematic of the method.

relative phase of successive laser pulses incremented by a fixed amount so that the phase of the k^{th} laser pulse given by

$$\Delta\phi_k = 2\pi k/M \;. \tag{1}$$

The total deposition on the N-photon absorber is then given by

$$I(N,M) = \sum_{k=1}^{M} \left(E_k E_k^*\right)^N \tag{2}$$

with

$$E_k = e^{i\pi x/\chi} + e^{-i\pi x/\chi} e^{i\Delta\phi_k} \;. \tag{3}$$

Here $\chi = \lambda/2\sin\theta$ is the period of the (1-photon) intensity pattern created by the interference of the two beams, where θ is half their angular separation. To achieve minimum feature size, one would set θ equal to 90 degrees corresponding to the situation of two beams striking the recording plate at-near grazing incidence. However, in our experiment described below, we used a much smaller angle to allow us to study the nature of the enhanced resolution. If phase shifts $\Delta\phi_k$ were not introduced, the resulting excitation pattern would reduce simply to $I(N,M) = [1+\cos(2\pi x/\chi)]^N$. Such a pattern would have the same spatial period as a conventional 1-photon interference pattern, although with sharpened features.

The effect of averaging M laser shots with progressively increasing phase shifts is to average out the undesired, slowly spatially varying terms, leaving only a spatially uniform component, the $\cos(2\pi Mx/\chi)$ component at the desired frequency, and possibly harmonics of this component if N is at least twice as large as M. Thus, the pattern has a resolution M-times better than that allowed by normal interferometric lithography. The visibility of this pattern is readily found to be given by

$$V = \frac{A_{M,N} + \sum A_{MHo}}{A_{0,N} + \sum A_{MHe}} \;, \tag{4}$$

where

$$A_{k,N} = (2N)!/[(N-k)!(N+k)!] \quad (k \neq 0) \tag{5a}$$

and

$$A_{0,N} = (2N)!/[2(N!)^2] \tag{5b}$$

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with $A_{0,N}$ the dc component of the deposition pattern, $A_{M,N}$ the desired component, A_{MHe} the even harmonics of the desired frequency, and A_{MHo} the odd harmonics. Notice that the even harmonics act to slightly reduce the visibility as they have a maximum

Table 1. Visibility as a function of resolution and absorption process.

Resolution Enhancement (M)	2			3			4		
Order of Absorption (N)	2	3	6	3	6	8	4	6	8
Visibility (V)	33	60	94	10	48	67	03	14	28

where the fundamental is a minimum, while the odd harmonics slightly improve the visibility as they have minima where the fundamental is a minimum. For $M = N$, the visibility drops off rapidly with increasing resolution (M). In the specific case of $M = N$, a proposal of this type of technique has previously been introduced [12]. However, the current proposal introduces a great improvement by allowing and analyzing cases for which $N \neq M$. For $N >> M$ the visibility can be much higher, always approaching unity in the limit of large N. As an example, for a factor of two resolution enhancement (M = 2), the ideal visibility is 1/3 for N = 2, but increases to 3/5 for N = 3. Table 1 gives a summary of the visibility for various combinations of N and M.

The experimental set-up we used to demonstrate this effect is shown in Fig. 2. The output of a picosecond Nd:YAG laser (λ = 1064 nm, τ = 25 ps, f = 10 Hz) is directed onto a thin plate beamsplitter. The transmitted component propagates to a right-angle prism, where it is translated and reflected back to the beamsplitter, while the reflected component from the beamsplitter propagates to a plane mirror and is reflected back at an angle such that it will overlap the translated component in the detection plane. In practice, one would place an N-photon-absorbing lithographic plate in the detection plane. However, for the results presented here, we simulated the properties of an N-photon absorber by an N^{th}-harmonic generator followed by a CCD camera. Any light not at the N^{th}-harmonic frequency was spectrally filtered so that only the desired harmonic was recorded by the CCD. M shots were collected and summed by a computer.

The angle between the interfering beams is set by adjusting the amount of translation introduced by the prism and the distance from the beamsplitter to the detection plane. For this experiment, the beams are separated by approximately 4-5 mm at a distance of approximately 2.5 m, leading to a fundamental period of $\chi \approx 300\,\mu$m. The prism is mounted on a micrometer-controlled translation stage, which is used as the phase shifter.

Figure 3 shows examples of the raw image data. Notice that for M = 1 [that is, when the averaging of phase shifted laser shots is not utilized, as illustrated in panels (a), (b), and (c)], the fringe spacing remains constant as N is increased, the only change being a sharpening of the fringes with increasing order of nonlinearity N. However, when two or more phase-shifted exposures are averaged together (M = 2 or 3), a doubling or tripling of the resolution is seen.

The full widths at half maximum of the beams were on the order of 600 μm. The usual interference pattern one would record on a linear absorber (that is, $N = M = 1$) is shown in (a). As predicted, resolution doubling was obtained in (d) and (e) for which M = 2, while resolution tripling (M = 3) is seen in (f). Due to slight differences in the alignment of our apparatus for the various cases shown here, the fundamental period is slightly different for the various curves.

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The data for the case of $N = M = 2$ agrees well with the theory in both period and visibility. Most of the deviation is due to noise in the wings of the beams. For the case of $N = 3$ and $M = 2$, the data still agrees well with theory, but with a visibility slightly lower than that predicted theoretically. The origin of the lowered visibility is most likely the different efficiency experienced by different spatial frequency components in the harmonic generation process as a consequence of phase matching effects. This effect would alter their relative strength in the field expansion.

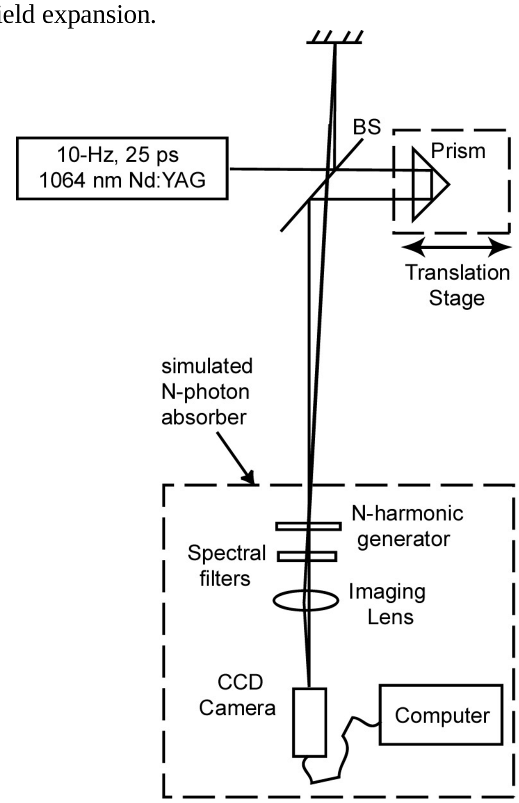

Fig. 2. Experimental set-up.

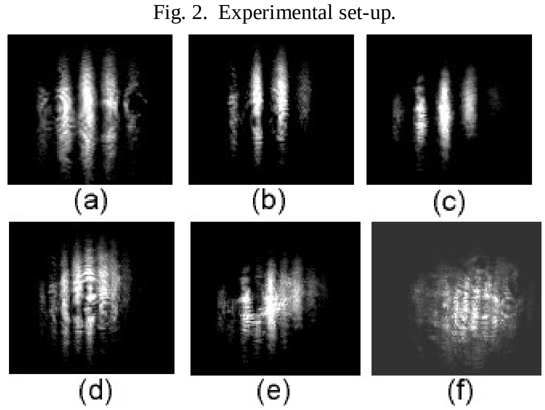

Fig. 3. Measured intensity distributions for (a) $M = N = 1$, (b) $M = 1$, $N = 2$, (c) $M = 1$, $N = 3$, (d) $M = N = 2$, (e) $M = 2$, $N = 3$, and (f) $M = N = 3$. Note that the first three patterns have the same period (because $M = 1$) but that the fringes become sharper with increasing N. Note also the doubling of the fundamental frequency in (d) and (e) and the tripling of the frequency in (f).

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Finally, for the case of $N = M = 3$, the data show a fringe spacing that is one-third of that for N=1. The fringe visibility is reduced as predicted by the theory. In addition to the noise sources encountered in (d) and (e), noise in (f) can arise from errors in the relative phase shifts. Since for $M > 2$, multiple phase shifts must be imposed, slight errors in the phase shift become more of a problem with increasing *M*. Also, for increasing M, each individual phase shift becomes smaller, which requires greater phase resolution. These problems could be overcome in a permanent set-up by using a mechanically rigid electrooptic device to perform the phase shifting.

The primary impediment to implementing this technique is the lack of suitable *N*-photon absorbing media, especially for *N* large. We note, however, that it is not crucial that the absorbers be ideal. If the absorbers are nonideal, in that is exhibit P-photon absorption (for $P < N$) as well as *N*-photon absorption, the only consequence would be a reduction in the visibility with no change in the resolution.

Another method for increasing the resolution of an optical lithographic system based only on the classical properties of light fields has recently been proposed [13] and a two-fold enhancement has been demonstrated through use of this technique [14]. This method entails the use beams with multiple frequency components, generated by means of a nonlinear mixing process. We feel that the method introduced in the present Letter possesses the same desirable features as this method but is much easier to implement. In addition, superresolution of spectral features using a multiphoton method has recently been reported, and the relationship between this method and quantum lithography has been noted [15].

Two final comments regarding the extension of this technique to more practical situations are in order. First, the wavelength used for the present experiment was determined by source and detector availability in our laboratory and not because of any fundamental wavelength limitation. In an actual system, a much shorter wavelength would be used. The only limitation on wavelength is the availability of appropriate optical components. Moreover, our technique can be straightforwardly generalized to be able to write an arbitrary pattern onto the lithographic plate. An arbitrary pattern can be synthesized by a superposition of various spatial frequency components, each of which can be written using our method. The maximum frequency component is limited by *N* and the chosen component determined by *M*. The details of how this procedure might be implemented have been described in the context of quantum lithography by [5] and [16]. The process described in this manuscript is aimed specifically at interferometric lithography, not projection lithography. With a fixed angle between the interfering beams, a simple Fourier decomposition could be used to write any periodic pattern. If the system is designed such that the interference angle can be varied such that the fundamental frequency can be varied over a continuous range, in principle one could also write aperiodic patterns.

In conclusion, we have proposed a new technique for achieving arbitrary resolution enhancement for optical lithography. We demonstrated the technique for up to a factor of three improvement. The procedure is quite straightforward to implement. High quality *N*-photon lithographic absorbers are all that are required to take this idea from the laboratory to the production line.

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