# Progress in Quantum Lithography

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## ABSTRACT

We review our progress toward implementing the proposal of Boto et al. (Phys. Rev. Lett. 85, 2733-2736, 2000) for using entangled photons to increase the spatial resolution of the process of interferometric photolithography beyond that of the standard Rayleigh limit.

**Keywords:** Entangled photons, lithography, quantum imaging

## 1. INTRODUCTION

Several years ago, the group of Jon Dowling[1] proposed the use of entangled photons to produce interferometric interferometry with a resolution exceeding the standard Rayleigh limit. This idea is illustrated in Fig. 1. A laser beam excites a nonlinear crystal and two entangled photons are emitted through the process of parametric down convesion. These fields are then brought together at a 50/50 beamsplitter. By the nature of quantum interference, these two photons can exit the beam splitter either both from the upper port or both from the lower port, but one never finds one photon in each output port. The quantum state of the field leaving the beam splitter can thus be represented as $|20, 02\rangle$, where the notation is such that $|N0, 0N\rangle = |N0\rangle + |0N\rangle$ and where $|n, m\rangle$ denotes a state in which $n$ photons are in the upper output port of the interferometer and $m$ photons are in the lower port. These states are often referred to simply as NOON states. The two beams leaving the beam splitter are then brought together on a recording medium that functions by means of two-photon absorption. Interference fringes are formed, not by classical interference, but by quantum interference. The probability amplitude for two-photon absorption with both photons coming from the upper arm interferes with the probability amplitude for two-photon absorption with both photons coming from the lower arm. Since any classical phase shift resulting from propagation shows up doubled in the two-photon probability amplitude, the resulting interference pattern has twice the fringe density as the classical interference pattern. In many ways, the enhanced resolution can be understood from the point of view that the de Broglie wave length of a quantum state comprised of two entangled photons is half the classical wavelength associated with either photon.[2,3]

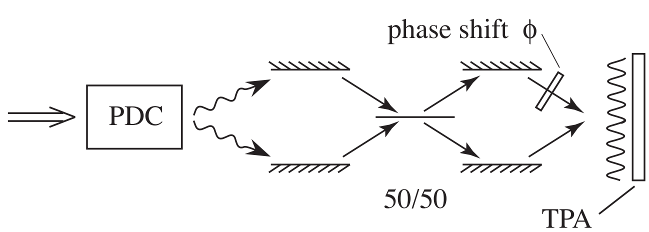


**Figure 1.** The idea of quantum lithography.

It should be noted that the quantum lithography process scales with with the number of entangled photons. In particular, if the $|N0, 0N\rangle$ state is incident on the beam splitter and the recording medium operates by $N$-photon absorption, the limiting resolution will be $\lambda/2N$.

While proof-of-principle studies of quantum lithography have been presented,[4,5] to date there have been no direct demonstrations of the quantum lithography process. There are two significant experimental challenges that must be met in order to demonstrate quantum lithography in the laboratory. First, one needs intense sources of photons in NOON states, and second one needs sensitive $N$-photon lithographic recording media. We next describe recent progress in these two areas.

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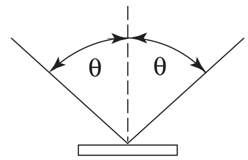


**Figure 2.** Experiemntal setup for writing gratings by multi-photon absorption in PMMA.

## 2. DEVELOPMENT OF INTENSE NOON STATES

According to the proposal of Boto et al.,[1] the quantum interference pattern is to be recorded by the process of $N$-photon absorption. Such multiphoton processes tend to be inefficient unless excited by intense optical fields. The challenge is to develop intense sources that possess the quantum statistics of NOON states. This challenge exists at both a practical level and at an in-principle level of finding interactions that will create NOON states at a high rate without allowing these states to overlap in time. If two NOON states were to arrive simultaneously at an $N$-photon absorber, it would be possible for the $M < N$ photons to be absorbed from one of these states and $N - M$ to be absorbed from the other. There is no guarantee that this absorption process would lead to the correct absorption rate to produce a fringe pattern of period $\lambda/(2N \cos\theta)$.

One possibility for producing an intense source of entangled photons for $N = 2$ is to replace the spontaneous parametric down-converter of the original proposal with a high-gain parametric amplifier that is seeded by spontaneous parametric down conversion. Such a device will clearly produce an intense output, but it is not immediately clear how the photon statistics will be influenced by the high gain of the process. It is well known, for instance, that all optical amplifiers add noise to the output beam. We have shown previously[6–8] by direct calculation that the consequence of using the intense output of a vacuum-seeded high-gain optical parametric amplifier is to decrease the fringe visibility of the resulting quantum interference pattern, while leaving the fringe spacing unchanged. Crucially, even in the limit of high gain, the fringe visibility never drops below 20%. We thus believe that this approach can lead to good results for quantum lithography with $N = 2$.

Quite recently[9] several new procedures have been reported for the production of entangled states with $N > 2$. For example, Mitchell et al.[10] have reported the construction of a $|30, 03\rangle$ state, and Walther et al.[11] have reported the construction of a $|40, 04\rangle$ state. Moreover, Eisenberg et al.[12] have reported evidence for entanglement of up to 100 photons. These are clearly very exciting developments, although the full implications for quantum lithography have yet to be fully explored.

## 3. DEVELOPMENT OF N-PHOTON LITHOGRAPHIC MATERIALS

The other experimental challenge is the development of an $N$-photon lithographic material. The approach that we have taken is to use poly methyl methacrylate (PMMA) as the lithographic material. We have selected this material because it is essentially transparent throughout the visible up to a wavelength of 250 nm. Our preliminary experiments were conducted using a Ti:sapphire laser operating at $\lambda = 800$ nm. Thus four-photon absorption is the lowest-order absorption process allowed for this setup. Because of the rapid decrease of excitation efficiency with increasing order of nonlinearity, we believe that four-photon absorption is the primary optical process leading to the results we have obtained.

The geometry used in our measurement is shown in Fig. 2. Each input beams stikes the sample at an angle of incidence of $\theta = 70$ degrees. The fundamental period of the interference pattern is thus $\lambda/(2\sin\theta) = 425$ nm. Each beam has the form of a pulse of 120 fs duration carrying 130 $\mu$J of energy, and is derived from the output of a Spectra Physics Spitfire regenerative amplifier. Our experimental procedure entails spin-coating a thin (approximately 2.5 $\mu$m thick) layer of PMMA onto a glass substrate. After exposure, we develop the plate for 10 sec in methyl isobutyl ketone (MIBK) and then rinse it for 30 sec in deionized water. We have analyzed the gratings formed in this manner using atomic force microscopy. Some of the results of this analysis are shown in Fig. 3. One can see especially from part (b) that the fringes are highly non-sinusoidal. This feature is a

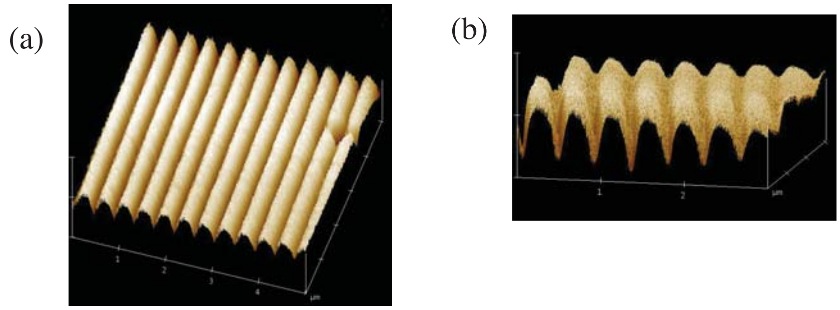


**Figure 3.** Atomic-force microscopy analysis of the fringes written by multiphoton absorption into PMMA.

consequence of the nonlinear nature of the multi-photon excitation process. The sharpness of the valleys of the fringes suggests that PMMA is capable of recording fringes with much smaller periods than those studied in the present experiment. The ability to record small-period fringes would be required for the demonstrations of sub-Rayleigh quantum lithography. It should be emphasized that the fringes reported by this work were produced by classical (not quantum) interferometry. The significance of the result is that it demonstrates that PMMA can serve as a high-resolution, multi-photon recording medium.

We also point out that there are some intriguing fundamental issues relating to multiphoton absorption excited by entangled photons. Both Javanainen and Gould[13] and Peřina et al.[14] have provided theoretical arguments showing that multiphoton absorption should scale linearly with intensity when excited by entangled photons. The argument is based on the observation that entangled photons would be expected to arrive simultaneously at an absorbing atom because they constitute a single quantum state. Experimental evidence for this behavior has been presented by Georgiades et al.[15] for two-photon excition of cesium atoms in a magneto-optic trap.

## 4. SUMMARY

In summary, we have presented an analysis of the current status of the field of quantum lithography. We believe that the intense field generated by an optical parametric amplifier gives the best tradeoff between possessing high intensity and the required quantum statistics for producing $N = 2$ sub-Rayleigh fringes. Recent experimental results on the formation of quantum states containing more than two entangled photons gives hope that quantum lithography with $N > 2$ may also soon be realizable. We have also presented the status our experimental work aimed at developing an $N$-photon lithographic material. It appears that PMMA is capable of serving as such a material with $N = 4$ and with a demonstrated spatial resolution of 425 nm.

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## REFERENCES

1. N. Boto, P. Kok, D. S. Abrams, S. L. Braunstein, C. P. Williams, and J. P. Dowling, Phys. Rev. Lett. 85, 2733-2736 (2000).
2. E. J. S. Fonseca, C. H. Monken, and S. Padua, Phys. Rev. Lett. 82, 2868-2871 (1999).
3. 5. K. Edamatsu, R. Shimizu, and T. Itoh, Phys. Rev. Lett. 89, 213601 (2002).
4. M. D'Angelo, M. V. Chekhova, and Y. Shih, Phys. Rev. Lett. 87, 013602 (2001).
5. S. J. Bentley and R. W. Boyd, Optics Express, 12 5735 (2004).
6. G. S. Agarwal, R. W. Boyd, E. M. Nagasako, and S. J. Bentley, Phys. Rev. Lett. 86, 1389 (2001).
7. E. M. Nagasako, S. J. Bentley and R. W. Boyd, and G. S. Agarwal, Phys. Rev. A 64, 043802 (2001).
8. E. M. Nagasako, S. J. Bentley and R. W. Boyd, and G. S. Agarwal, J. Mod. Opt. 49, 529-537 (2002).

9. D. Bouwmeester, Nature 429, 139 (2004).
10. M. W. Mitchell, J. S. Lundeen, and A. M. Steinberg, Nature, 429 161 (2004).
11. P. Walther et al., Nature 429, 158 (2004).
12. H. S. Eisenberg, G. Khoury, G. A. Durkin, C. Simon, and D. Bouwmeester, Phys. Rev. Lett. 93, 193901 (2005).
13. J. Javanainen, P. L. Gould, Phys. Rev. A 41, 5088 (1990).
14. J. Peřina, Jr., B. E. A. Saleh, M. C. Teich, Phys. Rev. A 57 3972 (1998).
15. N.P. Georgiades, E.S. Polzik, K. Edamatsu, H.J. Kimble, and A. S. Parkins, Rev. Rev. Lett. 75, 3426 (1995).