# **Quantum Imaging: New Methods and Applications**

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# **Quantum Lithography**

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# Demonstration of Sub-Rayleigh Lithography Using a Multi-Photon Absorber

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# **Original Quantum Lithography Proposal**



- Entangled photons produced in SPDC can increase resolution of an interferometric lithography system by factor of 2 (Boto et al., 2000)
- *N*-fold enhancement possible when *N* photons are entangled

Boto et al., PRL **85**, 2733 (2000)





# **Experimental Challenges**

- What quantum state of light to use?
  - Need <u>strong</u> enough light to excite two-photon absorption
  - Need <u>weak</u> enough light so that the statistics are those of individual photon pairs
- Develop a multi-photon absorber
  - N<sup>th</sup> harmonic generation/coincidence circuitry
  - Polymethylmethacrylate (PMMA)
    - Multi-photon absorber at visible wavelengths
    - e-beam resist





# Quantum Lithography with an OPA

- Replace parametric down-converter with high gain optical parametric amplifier (OPA)
  - Can be very intense
  - Possesses strong quantum features





Agarwal, Boyd, Nagasko, Bentley, PRL 86, 1389 (2001)



# **Experimental Setup**

#### Experiment to realize NOON (N=2) state for quantum lithography



**Problem:** Scattering from pump laser saturates detection with APDs  $\rightarrow$  Need to shield beam path.

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# **Two-Photon Excitation Rate**



For light from an OPA, both linear and quadratic dependence are present.

Cross-over point:

 $I = \frac{1}{3}$  photons/mode G = 0.55

For cases of practical interest, the rate scales quadratically with *I*.

Accepted for publication in JOSA B

# Visibility using an OPA and TPA





# Effect of an N-Photon Absorber

### (But still for a two-photon entangled state)



• As *N* increases the visibility improves, but there is no improvement in resolution.

Accepted for publication in JOSA B

# Summary of OPA Results

For most cases, two-photon excitation rate scales as *I*<sup>2</sup>.

- OPA + TPA produces fringes with visibility greater than 20%
- OPA + N-photon absorber produces fringes with even greater visibility (but with no greater resolution)



# Classically Simulated Quantum Lithography

## Proof-of-Principle Experiment

Bentley and Boyd, Opt. Exp. 12, 5735 (2004)



One-photon detector

Two-photon detector and one laser pulse

Two-photon detector and two pulses with phase shift







• Use classically simulated quantum lithography to develop an *N*-photon recording material

## **PMMA**

•Polymethylmethacrylate (PMMA) is a positive photo-resist that is transparent in the visible region.

•3PA @ 800 nm can break chemical bonds, and the affected regions can be removed in the development process.



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# **PMMA** Preparation

- Sample
  - PMMA (120,000 MW) + Toluene Solution (20% solids by weight)
  - PMMA is spin-coated on a glass substrate
    - spin-coated @ 1000 rpm, 20 sec
    - dried for 3 min
    - repeated 3 times
      - $\rightarrow$  1- $\mu$ m-thick film
- Development
  - Developer: 10 sec in 1:1 methyl isobutyl ketone (MIBK) to isopropyl alcohol
  - Rinse: 10 sec in DI water
  - Air blow dried



# **Experimental Setup**



WP: half-wave plate; Pol.: polarizer; M1,M2,M3: mirrors; BS: beamsplitter; f1,f2: lenses; PR: phase retarder (Babinet-Soleil compensator)



# Fringes on PMMA



Recording wavelength: Pulse energy: Pulse duration: Recording Angle θ:

Period: 425 nm

800 nm 130 μJ/beam 120 fs 70°

### Surface Cross-Section





### AFM images of PMMA surface





# Sub-Rayleigh Fringes on PMMA



AFM image of PMMA surface

#### Two pulses with $\pi$ phase-shift

Recording wavelength:
Pulse energy:
Pulse duration:
Recording Angle $\theta$ :

800 nm 90 μJ/beam 120 fs 70°

#### Period: 213 nm





# **Further Enhancement?**

- PMMA is a 3PA @ 800 nm, so further enhancement should be possible.
- Illuminate with two pulses with a  $2\pi/3$  phase-shift.





## 1/6 the recording wavelength!



# Importance of PMMA Result

- Demonstrates sub-Rayleigh resolution on a real material using the phase-shifted grating method.
- Shows that PMMA is an *N*-photon absorber with adequate resolution for use in true quantum lithography.
- But we are looking into other materials that can provide greater sensitivity.

## **Non-sinusoidal Patterns**

- In principle, Fourier's Theorem can be applied to generate arbitrary patterns.
  - Can only remove material
  - Visibility???
- Alternatively, we can generalize method...

$$E_{N,M} = \sum_{k=1}^{M} I_k^N \quad \text{where} \quad I = \left| 1 + A_k e^{i\chi} e^{i2\pi k/M} \right|^2$$

New term: Allow different amplitudes on each shot



# Non-Sinusoidal Patterns - Theory

 Different field amplitudes on each shot can generate more general non-sinusoidal patterns.

$$I = \sum_{m=1}^{M} A_m [1 + \cos(Kx + \Delta_m)]^N$$



For example, if N = 3, M = 3

$$A_1 = 1$$
 $\Delta_1 = 0$  $A_2 = 0.75$  $\Delta_2 = \pi/2$  $A_3 = 0.4$  $\Delta_3 = \pi$ 





# **Two-Dimensional Patterns - Theory**

 Method can be extended into two dimensions using four recording beams.

Pattern = thickness – 
$$\sum_{mx,my=1}^{M} A_{mx} [1 + \cos(Kx + \Delta_{mx})]^{N} A_{my} [1 + \cos(Ky + \Delta_{my})]^{N}$$
  
For example,  
N=8, M=14

With S. Bentley, Adelphi University



- PMMA is a suitable multi-photon absorbing material for use in quantum lithography
- Demonstrated sub-Rayleigh ( $\lambda/4$ ) resolution using NL lithography

## Ongoing research

- Construct an intense light source based on high-gain optical parametric amplification
- Demonstrate quantum lithography



# Entanglement Propagation in Photon Pairs created by SPDC

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Presented at OSA Annual Meeting, October 10<sup>th</sup>, 2006



Entangled position and momentum spaces in SPDC involve high dimensional Hilbert spaces.

### Questions:

- Effects of free-space propagation on the spatial correlations between photons?
- Implications about the entanglement present in the system?
- How rapidly do these correlations degrade as a consequence of loss or propagation through turbulence?

# **Spontaneous Parametric Down-Conversion**



$$\mathbf{k}_{\mathrm{p}}^{(\mathrm{e})} = \mathbf{k}_{\mathrm{s}}^{(\mathrm{o})} + \mathbf{k}_{\mathrm{i}}^{(\mathrm{e})}$$





## Propagation of Correlations



Question: What is going on in the intermediate zone? Is entanglement "lost"?

# **Experimental Set-up**



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# **Near-field Correlations**

 $( \underbrace{\mathsf{u}}_{i} \underbrace$ 

Normalized Coincidence Rates

- Slits/knife edge placed in the imaged plane of the crystal.
- Strongly correlated photon positions.

•

![](_page_28_Picture_5.jpeg)

# **Far-field Correlations**

#### Normalized Coincidence Rates

![](_page_29_Figure_2.jpeg)

- Slits/knife edge placed in the focal plane of the lens.
- Strongly anti-correlated photon positions.
- R<sub>x</sub> = 28.52

![](_page_29_Picture_6.jpeg)

# The Intermediate Zone?

![](_page_30_Figure_1.jpeg)

- Slits moved behind image plane (12 cm behind image plane or 14.5 cm after crystal).
- Little residual spatial correlations.

![](_page_30_Picture_5.jpeg)

# Migration of Entanglement to Phase

 At a particular propagation distance z<sub>0</sub> the amplitudesquared wave function will become separable, although the wave function itself is entangled, i.e.

$$|\psi(x_s, x_i)|^2 = f(x_s)g(x_i)$$
  
$$\psi(x_s, x_i) = \sqrt{f(x_s)g(x_i)}e^{i\phi(x_s, x_i)}$$

• The entanglement has migrated to phase. We need interferometric methods to get information about the entanglement.

$$|\psi(x_s, x_i) + \psi(x_s, -x_i)|^2$$

![](_page_31_Picture_5.jpeg)

# **Experiment to Detect Phase Entanglement**

![](_page_32_Figure_1.jpeg)

![](_page_32_Picture_2.jpeg)

# Conclusions

- We have experimentally investigated the propagation of spatial correlations in SPDC.
- The apparent disappearance of entanglement can be explained by the migration of entanglement from intensity to the phase of the wave function.
- Rotational shearing interferometers should enable us to recover the entanglement information in the intermediate zones.

![](_page_33_Picture_4.jpeg)

## Quantum Imaging: New Methods and Applications Quantum Lithography and Large Entanglement

#### **Objectives**

- Develop sub-Rayleigh photo-lithography
- Develop related imaging modalities
- Exploit entanglement in large Hilbert spaces for imaging applications
- Develop means of minimizing the loss of quantum coherence due to propagation

![](_page_34_Picture_6.jpeg)

#### Approach

- Develop sensitive multiphoton lithographic materials
- Make use of high-gain OPA as source of light for quantum lithography
- Exploit continuous-variable entanglement to produce high-dimensional entanglement
- Develop imaging protocols based on highdimensional entanglement

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#### Accomplishments

- Established PMMA as a suitable lithographic material
- Demonstrated ability to write  $\lambda$ /6 features
- Developed protocol for writing non-sinusoidal features
- Performed study of properties of OPA as a source for quantum lithography
- Performed measurements of modification of entanglement upon propagation

# Acknowledgements

![](_page_35_Picture_1.jpeg)

Supported by - the US Army Research Office through a MURI grant

![](_page_35_Picture_3.jpeg)

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