Quantum interference fringes beating the diffraction limit

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Abstract: Spatially formed two-photon interference fringes with fringe periods smaller than the diffraction limit are demonstrated. In the experiment, a fringe formed by two-photon NOON states with wavelength λ=702.2 nm is observed using a specially developed near-field scanning optical microscope probe and two-photon detection setup. The observed fringe period of 328.2 nm is well below the diffraction limit (351 nm = $\lambda/2$). Another experiment with a path-length difference larger than the coherent length of photons confirms that the observed fringe is due to two-photon interference.

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1. Introduction

Optical lithography is an indispensable technique for the mass production of microstructures such as semiconductor devices. However, there exists a resolution limit: the minimum period of the interference fringe is λ/2 for conventional light sources [1]. As the design rules of integrated circuits get smaller, the diffraction limit becomes a big problem. Quantum lithography [2-6] is a method to overcome this limit completely, since the resolution limit for an N-photon entangled state is reduced to λ/2N. Several experiments have observed a reduced de Broglie wavelength of photons in the time domain [7-9]. In the spatial domain, the reduction of the double-slit interference diffraction pattern by a factor of 2 using two-photon interference has been reported [5, 6]. However, the direct observation of spatially-formed interference-fringe periods smaller than λ/2 in the spatial domain, which is an indispensable step for the realization of quantum lithography, has not been reported. Here we report the direct observation of interference fringes beating the diffraction limit using a high-fidelity entangled photon source [10], a stable interferometer without an active stabilizer [11], and a specially developed near-field scanning optical microscope (NSOM) probe [12]. Our result confirms quantum lithography as a viable technique and opens up new possibilities in the use of quantum-optical phenomena in nano-technology.

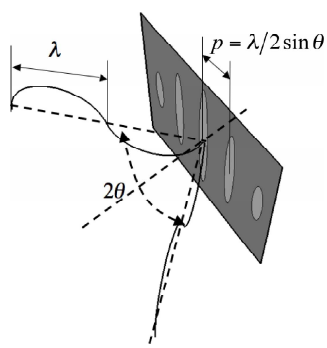

Fig. 1. Interference of two coherent plane waves. When two coherent plane waves with wavelength λ intersect at an angle of 2θ, an interference fringe with period p (= $\lambda/2sin\theta$) is obtained. An interference fringe modified by a limited beam diameter is shown for comparison with experimental results.

When two coherent plane waves with wavelength λ intersect at an angle of 2θ (incidence angle θ), the intensity profile of the laser shows fringes due to interference: $I = I_0\left(1+cos(2\pi r/p)\right)$, with a fringe period $p = \lambda/2sin\theta$ and where r is the position along the intersecting plane (Fig. 1). The period can never be smaller than $p_{min} = \lambda/2$, which is realized when $\theta = \pi/2$. This is called the Rayleigh diffraction limit and is the best fringe resolution that can be achieved classically. However, quantum lithography [2] can overcome this limit by using the quantum nature of entangled photons. Consider N entangled photons in

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spatially different optical modes (optical paths): $|\psi\rangle = \left(|N\rangle_\alpha |0\rangle_\beta + |0\rangle_\alpha |N\rangle_\beta\right)/\sqrt{2}$ as an input state. In this case, the probability to find N photons at a position r is given by

$$I_N = \{1 + cos(2\pi Nr/p)\}/2, \tag{1}$$

which exhibits a fringe period reduced by a factor of $1/N$. By using a multi-photon absorbing photo-resist material, it is possible in principle to use this phenomenon to improve the resolution of the lithography.

Note that the fringe peaks can be narrowed using only two-photon absorbing material and a classical light source, however, the period of the fringe does not change in this case [2]. As a related technology, two-photon interference with a thermal light field has been investigated [13,14]. In this case, interference fringes with a reduced period are only observed by using detectors at different spatial points and hence the technology cannot be directly used for lithography. Note also that in our experiments, the NSOM probe is used as a high-resolution spatial filter and is not used to detect the near-field (evanescent field).

2. Experimental setup

As a first step to realize quantum lithography, it is important to observe multi-photon interference fringes beating the diffraction limit. The lack of 'two photon detectors' with a high efficiency and a high spatial resolution has been a significant problem. We thus constructed a two-photon detector using an NSOM probe [12], which is used as a sub-micron scale spatial filter. The two photon detector consists of a NSOM probe with a single mode fiber output which is divided by a beam splitter and monitored by two single-photon counting modules (SPCM). However, the small collection efficiency (typically 7.5×10^{-5}) of the conventional NSOM probe is insufficient for coincidence counting, for which the event detection probability is proportional to the square of the collection efficiency. We overcame this problem by using a specially developed NSOM probe with an elliptical opening (Fig. 2 inset, minor axis: 0.2 μm, major axis: 1.8 μm) based on a polarization NSOM probe (JASCO International Co., Ltd.). The collection efficiency was improved to 3.3×10^{-3} maintaining the sub-wavelength resolution along the axis perpendicular to the fringes, giving a $2.0\times10^{3} = (3.3\times10^{-3} / 7.5\times10^{-5})^2$ improvement in the coincidence rate.

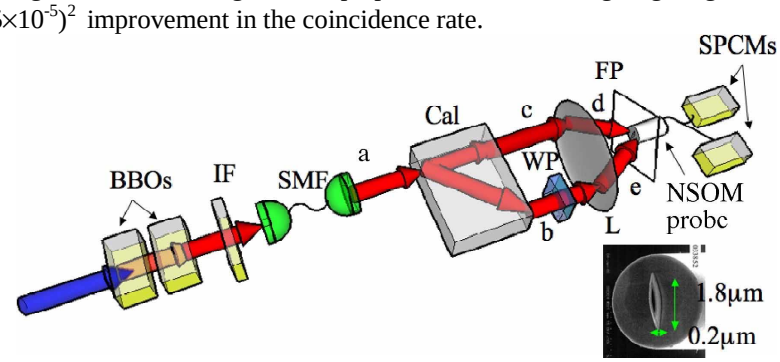

Fig. 2. Schematic of experimental setup. Photon pairs entangled in polarization, $\left(|VV\rangle_a + |HH\rangle_a\right)/\sqrt{2}$, are generated from two beta barium borate (BBO) crystals and pass through an interference filter (IF) and a single mode fiber (SMF). Then, the entangled photons are spatially separated into paths *b* and *c* depending on their polarization in a calcite crystal (Cal). The polarization in path *b* is rotated by 90 degrees by a half wave plate (WP), giving a two-photon NOON state ($\left(|2\rangle_d |0\rangle_e + |0\rangle_d |2\rangle_e\right)/\sqrt{2}$). An aspheric lens (L) is used to focus the photons to a small spot and to form an interference fringe in the focal plane (FP). An NSOM probe with an elliptical opening (inset) is scanned with a piezo-actuator along the focal plane. The single-mode fiber output of the probe is divided by a 50:50 fiber coupler and detected by single-photon counting modules (SPCM).

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The experimental setup is shown in Fig. 2. A single-frequency laser beam with wavelength 351.1 nm is incident on two beta barium borate (BBO) crystals for the type-I collinear phase matching condition. The optical axes of the crystals are set orthogonal to each other and diagonal to the polarization of the pump laser beam in order to generate a polarization entangled state, $\left(|VV\rangle_a + |HH\rangle_a\right)/\sqrt{2}$, via spontaneous parametric down conversion (SPDC) [10]. The polarization-entangled photons pass through an interference filter (IF; center wavelength 702.2 nm, FWHM 10 nm) and a single mode fiber (SMF). The change of the polarization in the SMF is analyzed using quantum-state tomography for the photons at the output of the SMF and compensated using a half wave plate and a quarter wave plate (not shown in Fig. 2). The compensated two-photon entangled state in polarization is transformed by a calcite crystal (Cal) into two paths, *b* and *c*, according to the polarization [11], $\left(|VV\rangle_b + |HH\rangle_c\right)/\sqrt{2}$. Then, the state is changed into a two-photon NOON state [15,16], $\left(|2\rangle_d|0\rangle_e + |0\rangle_d|2\rangle_e\right)/\sqrt{2}$ by rotating the polarization in path *b* by 90 degrees. A coupling lens with a large numerical aperture (L; clear aperture = 3.6 mm, NA = 0.65) is used to superpose the two modes with a large intersecting angle 2θ to give an interference fringe of small period (Eq. (1)). Since the two photons in a pair pass through the same optical components, the fluctuation of the path-length difference is minimized and results in super-stable interference fringes, which remain unchanged for about ten hours. Then, an NSOM probe (inset) is scanned with a piezo-actuator along the focal plane. The single-mode fiber output of the probe is divided by a 50:50 fiber coupler and detected by single-photon counting modules (SPCM-AQR14FC, PerkinElmer). The coincidence events between the two SPCMs are recorded by a gated photon counter (SR-400, Stanford Research Systems).

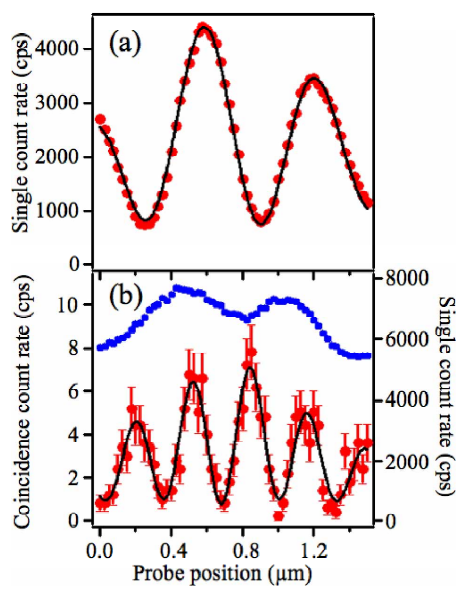

Fig. 3. (a) Interference fringe of single photons. The lateral axis indicates the position of the NSOM probe. The vertical axes indicate single counting rates. A polarizer is placed before the single mode fiber to select only horizontally polarized photons. The polarization is then rotated 45 degrees. Since the effective detection efficiency ($<1\times10^{-3}$) including the collection efficiency of the probe is so small, the generated fringe is the same as that for a single-photon state in two modes, $\left(|1\rangle_d|0\rangle_e + |0\rangle_d|1\rangle_e\right)/\sqrt{2}$, when only the output of one of the two detectors is recorded. The black line is a sinusoidal curve weighted by a Gaussian function

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(see text) fitted to the experimental data. (b) Interference fringe of entangled photons beating the diffraction limit. The vertical axes indicate coincidence count rates (red dots, left axis). The black line is a fit to the experimental data. The fringe period of 328.2 nm is smaller than the diffraction limit of 351.1 nm. Single count rates of one of the two detectors measured at the same time are shown for reference (blue dots, right axis). For (a) and (b), the NSOM probe was scanned with 25-nm steps. The accumulation time for one data point was 5 seconds. The error bars show $\pm\sqrt{counts}$ assuming Poisson statistics.

3. Results and discussion

Figure 3(a) shows an interference fringe of single photons taken for reference (see figure caption). The experimental data (red dots) is fitted well with a theoretical curve (black line), consisting of a sinusoidal curve weighted by the Gaussian distribution of the incident spatial mode profile (beam diameter 4.14 μm estimated from Fig. 4(a)). The period of 656.4 ± 3.8 nm estimated from the fit to the experimental data is smaller than the wavelength of the photon (702 ± 5 nm), indicating that with two-photon interference it will be possible to beat the diffraction limit. The intersection angle 2θ of the incident beams was estimated to be 65.4°, which agrees with the value estimated for the beam-injection setup.

The key experimental result of an interference fringe beating the diffraction limit is shown in Fig. 3(b) (red dots). It is clear that the fringe period of Fig. 3(b) is half that of the classical case (Fig. 3(a)). The experimental data are well fitted with a theoretical curve (black line) with a period of 328.2 nm = 656.4 nm / 2, which is well below the diffraction limit of 351.1 nm. The figure also shows a high fringe visibility $(=(I_{max}-I_{min})/(I_{max}+I_{min}))$ of 70.7± 7.8%. It should be mentioned that for such a high-visibility interference fringe, considerable work was required to improve the quality of the entangled photons incident on the calcite crystal via a single-mode fiber (shown in Fig. 2) using polarization compensators (not shown in Fig. 2). The data shown in Figs. 3(a) and 3(b) were measured by scanning the probe position from 0 μm to 1.5 μm (single scan), changing the setup for single-count data (Fig. 3(a)) and coincidence data (Fig. 3(b)) using computer-controlled motorized stages. For reference, single count rates of one of the two detectors measured at the same time with the coincidence count rate are shown as blue dots. A slight modulation (about 10% on average) with a period of about 656 nm is observed, which may be caused by single photon interference of the two-photon source, which has still imperfection compared to an ideal source.

We can confirm that the result shown in Fig. 3 is due to the quantum interference of two photons by a similar method as that performed for measuring the de Broglie wavelength [9]. Since the photon pairs were generated via an SPDC process, the photons were entangled in time or in frequency. Thus, the effective coherence length of two-photon quantum interference is not given by the coherence of each single photon but by the coherence of the pump laser. Hence, two-photon interference can be observed even when the path difference is much larger than the coherent length of each photon [9].

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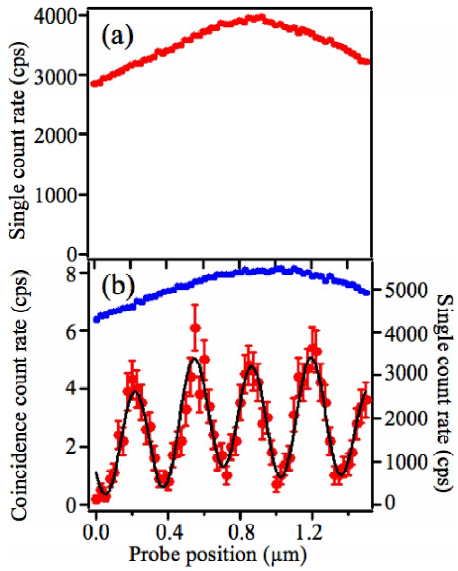

Fig. 4. Interference fringes of (a) single photons and (b) entangled photons for a path difference much larger than the coherent length of each photon (~75 μm) (red dots, left axis). The path difference of about 500 μm was achieved by inserting a glass plate in one of the optical paths. The fringe period of 327.5 nm in (b) beats the diffraction limit of 351.1 nm. Single count rates of one of the two detectors measured at the same time are shown for reference (blue dots, right axis). The NSOM probe was scanned with a 25-nm step. The accumulation time for one data point was 10 seconds. The error bars show $\pm\sqrt{counts}$ assuming Poisson statistics.

Figure 4 shows the interference fringes when one of the effective path lengths was extended by about 500 μm by inserting a thin glass plate. Since the path length difference is much larger than the coherent length of the photons (75 μm, determined by a band-pass filter set in the entangled photon source), no interference can be observed for the classical case (single-photon counting, Fig. 4(a)). In contrast, an interference fringe with reduced period beating the diffraction limit is still observable when two-photon coincidence counts are measured, as shown in Fig. 4(b) (red dots). This experiment clearly demonstrates that the interference fringes shown in Figs. 3(b) and 4(b) are due to quantum interference, not a classical effect. This result also suggests an interesting feature of quantum lithography, namely that it can be very robust against path-length difference mismatch when auxiliary entanglement of photons in other degrees of freedom is used.

4. Conclusions

We believe the observation of interference fringes beating the Rayleigh diffraction limit opens new possibilities for quantum lithography. The next challenge is the fabrication of patterns beating the diffraction limit using real multi-photon absorbing photo-resist material. In addition to the experimental technologies developed here, ultra-bright sources of entangled photons [17,18] and multi-photon absorbing photo-resist materials [19] with huge cross-section may be used for such an experiment. Note also that the resolution of quantum lithography can be dramatically improved when the number of entangled photons is increased, since the diffraction limit for N-photon interference is not $\lambda/2$ but $\lambda/2N$ [2]. Recently, several experiments have observed reduced de Broglie wavelengths of photons [8,9,20] using states with N=3 or N=4. The resolution of quantum lithography using entangled four photons with a wavelength of 700 nm [8] is comparable with that of conventional optical lithography using deep-UV light (175 nm). Furthermore, when a 10-photon entangled state is used for the input, the limit becomes $\lambda/20$. This means that optical

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lithography with less than 30-nm design rule is possible using conventional optics for visible light.

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