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Quantum lithography: status of the field

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Abstract This contribution provides an analysis of progress in the field of quantum lithography. We review the conceptual foundations of this idea and the status of research aimed at implementing this idea in the laboratory. The selection of a highly sensitive recording material that functions by means of multiphoton absorption seems crucial to the success of the proposal of quantum lithography. This review thus devotes considerable attention to these materials considerations.

1 Background and conceptual issues involving quantum lithography

Quantum lithography is a technique first proposed by Boto et al. [1] that allows one to write interference fringes with a spacing N-times smaller than the classical Rayleigh limit of resolution, which is approximately $\lambda/2$. The basic idea of quantum lithography is illustrated in Fig. 1 for the case $N = 2$. Here a laser beam is allowed to fall onto a nonlinear mixing crystal in which parametric downconversion occurs, producing two daughter photons each at twice the wavelength of the pump laser beam. This photon pair then falls onto a 50-50 beam splitter. The output of the interferometer under these circumstances is the entangled state $|2, 0\rangle + |0, 2\rangle$, where the notation is such that

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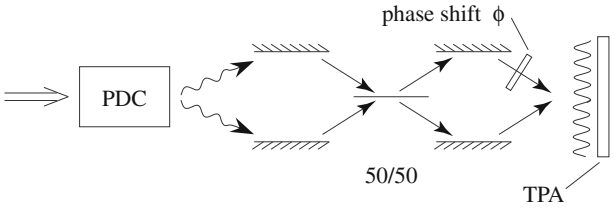

Fig. 1 Schematic experimental set-up for quantum lithography:*PDC* parametric downconverter. This set-up can be used to record interference fringes with a spacing that is twice as dense as that of classical interference

$|n, m\rangle$ denotes a state in which n photons are in the upper output port of the beamsplitter and m photons are in the lower port [2]. Note that, as a consequence of quantum interference, one never finds one photon in each output port. More generally, the state $|N, 0\rangle + |0, N\rangle$ is known as a N00N state. These two output beams then interfere on a recording medium that responds by means of two-photon absorption. Fringes are formed by means of the interference between the probability amplitudes for two-photon absorption with the photon pair taking either the upper or the lower pathway. Each of these probability amplitudes depends on path length L as exp*(2ikL)*, where the factor of 2 occurs because each of the two photons acquires the phase shift kL. The fringe spacing is thus twice as fine as that given by the normal interference patterns between the two waves. In many ways, the enhanced resolution can be understood from the point of view that the de Broglie wavelength of a quantum state consisting of two entangled photons is half the classical wavelength associated with either photon [3,4]. Figure 1 has been drawn with the beams striking the recording medium at oblique incidence, although in practice the beams would be directed towards the medium at near-grazing incidence to achieve the finest fringe spacing possible. Boto et al. showed that the ability to write small structures scales with the order N of the interaction. That is, if N photons are entangled and the recording medium responds by N-photon absorption, features of size $\lambda/(2N)$ can be written into the material.

The prediction of Boto et al. in 2001 generated great interest because of the enormous potential importance of the approach, and in fact soon thereafter a mockup proof-of-principle experiment by D'Angelo, et al. [5] was reported in the same year. (This experiment used two detectors operating in coincidence to mimic the effect of a true two-photon absorbing resist.) Nonetheless, if became almost immediately clear that a true laboratory verification of quantum lithography would be extremely challenging. In particular, implementation would require a lithographic material that would respond by means of N-photon absorption excited by quantum states in the form of N00N states, which are expected to possess a very small photon flux.

Much research over the past decade has been aimed at developing intense sources of quantum states of light such as N00N states [6]. One suggestion for generating an intense beam of N00N states for $N = 2$ is to generate the quantum states of light by means of an unseeded high-gain optical parametric amplifier (UHGOPA) instead of using a spontaneous parametric down converter (SPDC) as assumed in the original Boto proposal. Detailed theoretical treatments [7–12] have shown that even the intense output of such a device will possess quantum features strong enough to allow the

writing of high-contrast fringes in a quantum lithography configuration. It should be noted that the only difference between a SPDC and a UHGOPA is that the latter device is pumped much harder. Because it is pumped much harder, its output consists not only of biphotons but also of ensembles of biphotons. There is no guarantee that such a source will produce a N00N state or indeed even a linear superposition of N00N states for a range of N. Nonetheless, these theoretical treatments have shown that the sub-Rayleigh fringes can be produced through use of an UHGOPA as a light source, although the visibility of the fringes is reduced. Recent experimental work by one of us in collaboration with the group of DeMartini in Rome establishes the potential usefulness of an UHGOPA for quantum lithography [13]. However this experiment, similar to that of D'Angelo, et al. [5], was also carried out with coincidence counters mocked up to act like N-photon absorbers—no true N-photon absorbing material was used.

These theoretical and experimental results indicate that if we use the UHGOPA in conjunction with an N-photon absorber, we could increase the contrast and the fringe sharpness but not the fringe density beyond a factor of two. This is due to the fact that the dominant N00N output of the UHGOPA is a N00N state of $N = 2$. The other higher N00N components are essentially washed out due to the competition between different photon numbers in the wave function for the components with $N > 2$.

There has also been considerable interest in the question of how the efficiency of N-photon absorption depends on the entangled nature of the incident light beam. It has been known for some time [14–16] that the transition rate for two-photon absorption (TPA) can scale linearly (not quadratically) with laser intensity. This fact can be used to advantage in designing a quantum lithographic system, because the linear dependence is expected to dominate over the quadratic term under the low intensity conditions expected to be relevant for entangled light sources. In fact, several laboratory investigations have confirmed the existence of this linear dependence of the rate of two-photon absorption. These results are reviewed towards the end of the present section.

There has also been much discussion of how the spatial correlations of entangled photons can influence the efficiency of the quantum lithography process. In their original paper, Boto et al. [1] argue that quantum correlations will cause the two photons of a biphoton pair to always arrive at the same detector element, leading to a great enhancement of the efficiency of the process of quantum lithography. Subsequently, several authors [17–20] have argued that this analysis is incorrect, and that in fact if one photon falls onto a detector element its entangled partner is no more likely to land there than anywhere else. Two slightly different versions of this conclusion have been advanced. Tsang [18,19] points out that when one photon is localized, the momentum (wavevector) of the other photon become completely delocalized, and thus this photon can end up anywhere. In contrast, it has been argued [20] that the diffraction of the second photon is uninfluenced by the localization of the first. At present, it is not clear whether these two interpretations are at odds with one another or whether they in fact lead to identical predictions. Reference [20] has also pointed out that recent experimental results of Peeters et al. [21] demonstrate that the localization of the first photon does not lead to the second photon being localized to the same location. This conclusion, if correct, would imply great difficulties in implementing the Boto et al. [1]

version of quantum lithography because integration times would likely become excessively long. However, Plick et al. [22] point out that these conclusions hold only for a conceptually unrealistic idea of a plane wave N00N state, which is delocalized over all space, and that in fact a N00N state created by the laboratory process of SPDC can very well possess an enhanced localized two-photon detection rate, once the actual spatio-temporal properties of the wavefunction are properly analyzed. At the present, these issues are still being sorted out. The conclusion seems to be that the implementation of quantum lithography may not be quite as straightforward as the procedure proposed by Boto et al. but that nonetheless there may be means for increasing the efficiency of the quantum lithography process.

Much progress in super-resolved quantum imaging can be made, however, if the requirement for lithography and the N-photon resist are dropped. Tsang [23] has recently suggested a variation on the scheme of Boto, et al., that is expected to show much greater recording efficiency (that is, reduced integration time). In this scheme N entangled photons are allowed to fall onto a detector array, as opposed to an N-photon absorbing resist. For each measurement, the centroid of the N detected photo events is measured, and the probability distribution (histogram) of the centroid position is determined through repeated measurements. Tsang has shown that this probability distribution will show sub-Rayleigh features in the post-processing of the image. While this method does not lend itself to the recording onto a lithographic recording material, it could prove very useful in optical metrology and imaging.

A great deal of work by the Scully and Zubairy groups at Texas A&M University has gone into the investigation of non-traditional N-photon absorption schemes that are based on coherence effects in three or higher-level systems, including dopplerons. A recent example of this approach is in Ref. [24]. While this work has been very stimulating in the venue of the investigation of the quantum control of the absorption of nonclassical light in N-level atoms, it is difficult at present to see how such results may be translated to a practical N-photon absorbing resist for commercial use. However the work may find application in quantum imaging of biological samples tagged with specific atoms that have the desired properties as required by the Scully and Zubairy teams.

One of the concerns with the original work of Boto, et al., was that this initial scheme applied to the writing of absolutely periodic one-dimensional patterns. To be useful, an arbitrary pattern would need to be made. Two solutions to the arbitrary pattern problem appeared nearly simultaneously. The first, also by the Dowling group at JPL [25], showed that arbitrary patterns could be made in a few writings by decomposing the patterns into two-dimensional Fourier-like decompositions in the photon field. The drawback of the scheme was that to work it required the ability to make superpostions of N00N states of varying N in a two-dimensional array with various phase relations, which would put extraordinary constraints on the control and flux of the entangled light source.

An alternative scheme for arbitrary pattern drawing was proposed by Bjork et al. [26] in which the idea was to use N00N states of fixed N to write a sub-Rayleigh small spot and then to raster scan such spots to construct an entire image. While avoiding the requirement of creating superpositions of N00N states, the Bjork scheme has the disadvantage that it would be very slow, taking a long time to make each image,

rendering it less useful for high throughput commercial lithography applications. Also similar or even better resolution than this approach of Bjork et al., can be achieved by raster scanning an illuminated narrow fiber optic tip, a method known as near-field scanning optical lithography, where the evanescent field is not subject to the Rayleigh criteria, as we are now in the near field, and where no quantum state of the field is required at all to get good performance. This nonlinear optical classical approach is also slow and less practical than "single shot" lithography in the image plane [27].

2 Experimental studies of quantum lithography

As we mentioned in Sect. 1, it has been predicted theoretically [14,15] that when two-photon absorption is excited by entangled light fields the transition rate will scale linearly with the light intensity. This linear dependence is important for two different reasons. One is that a linear dependence will increase more rapidly than a quadratic dependence for weak light fields. The other is that a linear dependence will guarantee that the exciting light field possesses the proper sort of quantum correlations to produce quantum lithography. This prediction had been experimentally verified previously by the group of Kimble [16]. More recent demonstrations include those of Strekalov et al. [28], Dayan et al. [29], and Sensarn et al. [30].

We mentioned above that D'Angelo et al. [5] have performed a proof-of-principle experiment for the case $N = 2$ in which the properties of the two-photon recording medium were mimicked by an electronic coincidence circuit. 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.

Bentley and Boyd [31,32] have described 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 was demonstrated 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 was simulated by Nth 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.

Using a different procedure, Chang et al. [33] demonstrated the ability to use PMMA as a multi-photon recording material when excited by intense classical light. Because of the highly nonlinear nature of the recording process, a sinusoidal intensity distribution produced by normal interference gave rise to greatly narrowed recorded fringes. By adjusting the phase of one of the interfering beams between successive laser shots, it was possible to record a fringe pattern with a fringe density exceeding the classical Rayleigh limit. By means of this phase-shifted-grating method and a classical light source, they demonstrated a two-fold enhancement in resolution by writing fringes

with a period of $\lambda/4$. These results suggest that PMMA can be useful as a nonlinear lithographic material for the realization of quantum lithography.

To summarize, it appears that no compelling laboratory demonstrations of quantum lithography have thus far been reported. Laboratory demonstrations to date have fallen into two categories. Some use quantum light sources but record the interference pattern electronically. In this case there is no lithographic record that is preserved. In another class of experiments, permanent fringes are registered in a recording material, but the light source used to write these fringes is purely classical in nature. Nonetheless, hope remains for finding the right combination of light source and recording material for which true quantum lithography can be demonstrated. In the next section we turn to possibilities afforded by the proper choice of recording materials.

3 Selection of material systems for quantum lithography

An important consideration for the development of quantum lithography is the choice of recording material. As mentioned above, an appropriate recording material would need to be sufficiently sensitive to respond by N-photon absorption when excited by quantum states of light, which tend to carry low photon flux.

It is useful to begin this discussion by considering the sensitivity of typical recording materials that are commonly used with classical light sources. Historically, most of these materials have been intended to function by means of single-photon absorption. Silver halide photographic plates are an example of such a recording medium. These materials are typically used with an energy fluence of 1 mJ/cm^2. Dichromated gelatin plates, which are often used for holographic data storage, have sensitivities of approximately 100 mJ/cm^2. State-of-the-art photopolymers have also been developed that possess much greater sensitivities. One such material [34] functions at an exposure level of 0.5 mJ/cm^2.

Sensitivities are much lower for multiphoton recording materials. One very successful example of multiphoton recording has been the direct laser writing of three-dimensional optical structures. Because of the highly nonlinear nature of the writing process, it is possible to write three-dimensional structures because only in the intense focal region of the laser beam is the intensity high enough to modify the optical properties of the recording material. Early work of this sort includes that of Kawata's group [35,36], which made use of two-photon induced photopolymerization in a commercially available photopolymer. Typical values of the laser parameters used in this work are as follows: laser wavelength of 790 nm, laser pulse duration of 200 fs, laser repetition rate of 76 MHz, and peak laser power of 50 kW. Typical exposure times were 2.5 s when focusing was accomplished by a lens with a numerical aperture (NA) of 0.88. Typical exposure levels were 18 J/μ m^2. Direct laser writing has been implemented more recently with great success by the group of Wegener [37] using the lithographic material SU-8 [38]. A great variety of structures have been fabricated though use of this approach, including photonic crystals containing complete photonic bandgaps, novel resonator designs using these structures, 3D chiral photonic crystals, and photonic quasicrystals. A very significant feature of this approach is that after a structure is fabricated it is possible to back-fill the voids in

the structure with another material, such as gold or silicon. In this way, structures with large dielectric contrast can be fabricated. Typical parameters used in the direct laser writing are as follows: laser wavelength of 780–800 nm, laser pulse duration of 100–120 fs, laser repetition rates up to 1 kHz, and pulse energies of 9.5 nJ. The material was irradiated using an oil-immersion 100× microscope objective with an NA of 1.4.

Another approach to the storage of optical data in a dielectric medium involves the ablation of optical material or the permanent modification of the chemical structure of the material as the result of multiphoton ionization. For example, the group of Mazur [39] has demonstrated the ability to write permanent structures into glass using as few as 100 laser pulses each containing as little as 5 nJ of energy when focused with a lens with an NA of 1.4. The energy thus required for structural modification of this sort is thus as little as 500 nJ in a spot size of the order of 0.5 square micrometers. This group also showed that it is possible to write waveguide structures into this material through use of a pulse train, with a repetition rate of 25 MHz in this particular study.

Self-organized arrays of nanogratings excited by femtosecond laser radiation were reported by Shimotsuma et al. [40]. Precise measurements of the intensity and pulse-length dependence of the ionization process that leads to the modification of the optical properties of dielectric materials have been presented by the group of Corkum [41,42]. Typical laser parameters used in these studies were pulse durations in the range of 50–1,200 fs and peak intensities in the range of 1–25 TW/cm^2. Work has also been reported [43,44] on the development of new materials with properties especially tuned to the writing of structures of use in photonics.

The multiphoton absorption properties of recording materials can be quantified at a microscopic level in terms of the N-photon absorption cross section. An extensive compilation of multiphoton absorption cross sections has been presented by He et al. [45]. In brief summary, this work shows that typical molecules possess two-photon absorption cross sections of the order of 1 GM, where 1 GM $= 10^{-50}$ cm^2 s/photon. Molecules with unusually large two-photon cross sections can possess values as large as 47,000 GM [46]. Other workers [47,48] have shown that it is possible to enhance multiphoton cross sections by a factor of nearly 1,000 by means of local-field enhancement by placing the molecular species in close proximity of metallic nanoparticles that produce a plasmonic resonance. Another approach to the enhancement of multiphoton cross sections has been described by Dolgaleva et al. [49], who show that local field effects can lead to the cascading of lower-order nonlinearities to produce large high-order nonlinearities.

Some extremely interesting results have recently been reported by the group of Goodson [50,51]. Specifically, this group has shown that certain porphyrin and thiophene dendrimers display extremely large cross sections for two-photon absorption when excited by entangled photons.

We next analyze some specific results presented in their paper on porphyrin dendrimers [50]. They model their results by arguing that the transition rate R scales with the photon flux φ as

$$R = \sigma_e \varphi + \sigma_r \varphi^2 \tag{1}$$

Here σ_e is the cross section for entangled two-photon absorption and σ_r is the cross section for normal two-photon absorption (TPA), in which the photons arrive at random moments of time. These cross sections are related by [51]

$$\sigma_e = \sigma_r / 2 A_e T_e. \tag{2}$$

Here A_e is the entanglement area. It is a measure of the transverse dimensions of the fourth-order coherence function, that is, the distance over which two photons remain entangled. Also, T_e is the entanglement time. It gives the maximum time separation of two entangled photons. These authors estimate values of $10^{-2}\,\mathrm{cm}^2$ for A_e and 200 fs for T_e for their experimental conditions. They also find that σ_e is approximately $10^{-17}\,\mathrm{cm}^2$. The abstract to this work reports that their measured value of σ_r determined under classical excitation is approximately $10^{-48}\,\mathrm{cm}^4\,\mathrm{s}^{-1}$. However, in the body of the paper this group reports that a value of $10^{-32}\,\mathrm{cm}^4\mathrm{s}^{-1}$ is implied by the data collected with a high flux of entangled photons. Standard models of multiphoton absorption hold that these two values should be equal. The authors conclude that the physics of multiphoton absorption processes in these materials appears to be very interesting although somewhat complex and is still being investigated.

In their work on thiophene dendrimers [51], this same group has found that the cross section for normal (random) TPA was as large as 1,100 GM and that the cross section for ETPA was as large as $3.7 \times 10^{-17}\,\mathrm{cm}^2$. Both values were found to increase with the number of thiophene units contained in the dendrimer for values in the range of 6–90. They find that the one- and two-photon rates become comparable at a photon flux density of 3×10^{12} photons $\mathrm{cm}^{-2}\,\mathrm{s}^{-1}$. Excitation rates have to be kept below this level to ensure that only quantum-correlated photons contribute to the excitation process. This group has suggested that the cross section for ordinary two-photon absorption (TPA) does not necessarily scale in proportion to that for entangled two-photon absorption (ETPA). This observation suggests that the optimization strategies in selecting materials for the two cases can be quite different. The nature of absorption processes when excited by entangled photons has been investigated theoretically by Fei et al. [52].

In order to make use of ETPA for quantum lithography, one would need to find a means for writing permanent changes into the material following the excitation process. While it seems feasible to find the right combination of absorbers and activators that would allow for the permanent writing of these features, to date this capability has not been demonstrated.

4 Summary and conclusions

In summary, we have reviewed the status of the field of quantum lithography. The method holds great promise for the writing of sub-Rayleigh structures, and by extension to other sorts of sub-Rayleigh imaging. To date, no compelling demonstrations of quantum lithography have been presented, although proof-of-principle experiments that display certain aspects of quantum lithography have been presented. It is hope that with the development of new light sources and new lithographic materials it will be possible to implement true quantum lithography in the near future.

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