Quantum Lithography Has a Reduced Multiphoton Absorption Rate

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Abstract: It is shown that, contrary to popular belief, the multiphoton absorption rate is reduced if entangled photons are used to reduce the feature size of multiphoton lithography.

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One of the main promises of quantum lithography [1], besides the reduction of feature size, is the enhancement of the multiphoton absorption rate, since classical multiphoton lithography requires unrealistically high optical powers. Boto *et al.* argued heuristically that, for quantum lithography, as the photons are correlated in space, time, and number, they are contrained to arrive at the same place and the same time, leading to an enhancement of the multiphoton absorption probability. Here we show that, unfortunately, the heuristic argument by Boto *et al.* with respect to the spatial domain is not correct. In fact, the opposite is true: A reduction of feature size can only be achieved by spatially *anti-correlated* photons, as such, the photons arrive at the same place less often than uncorrelated photons, leading to a reduced multiphoton absorption probability. Consider the general N-photon state,

$$|\Psi\rangle = \frac{1}{\sqrt{N!}}\int dk_1 dk_2 ... dk_N \phi(k_1,k_2,...,k_N)|k_1,k_2,...,k_N\rangle = \frac{1}{\sqrt{N!}}\int dx_1 dx_2 ... dx_N \psi(x_1,x_2...,x_N)|x_1,x_2,...,x_N\rangle, \quad (1)$$

where ϕ is the configuration-space probability amplitude in the momentum domain and ψ is the corresponding quantity in the real space, related to the former by an N-dimensional Fourier transform. Both are subject to the normalization condition $\int dk_1 dk_2 ... dk_N |\phi|^2 = \int dx_1 dx_2 ... dx_N |\psi|^2 = 1$. The resolution limit means that $\phi(k_1,...,k_N) = 0$ for any $|k_i| > 2\pi/\lambda$. The N-photon absorption probability is given by $\langle : I^N(x) : \rangle = \frac{1}{N!}\langle\Psi|[\hat{A}^\dagger(x)]^N[\hat{A}(x)]^N|\Psi\rangle = |\psi(x,x...,x)|^2$, which is the *conditional* probability distribution *when all photons arrive at the same place x*. Hence, the multiphoton absorption rate depends on how often these photons arrive at the same place. The nonclassical state that achieves the minimum feature size is given by

$$\phi \propto \int dk G(k)\delta(k_1-k)\delta(k_2-k)...\delta(k_N-k), \quad \psi \propto \int \frac{dk}{\sqrt{2\pi}} G(k)\exp\left[iNk(x_1+x_2+...+x_N)\right], \quad (2)$$

$$\langle : I^N(x) : \rangle \propto \left|\int \frac{dk}{\sqrt{2\pi}} G(k)\exp(iNkx)\right|^2. \quad (3)$$

The original nonclassical state proposed by Boto *et al.*, $|\Psi\rangle \propto |N\rangle_{k'}|0\rangle_{-k'} + |0\rangle_{k'}|N\rangle_{-k'}$, is equivalent to the state given by Eqs. (2) when $G(k) \propto \delta(k-k') + \delta(k+k')$. Because all photons are constrained to have the same momentum, $G(k)$ is allowed to have the same resolution limit as the one-photon case, that is, $G(k) = 0$ for $|k| > 2\pi/\lambda$, thereby producing a minimum multiphoton absorption feature size $\sim \lambda/N$. However, the ideal state given by Eqs. (2) is not normalizable and therefore not physical. In fact, one can show that $\langle : I^N(x) : \rangle$ approaches zero if one constructs a normalizable state that approaches the ideal state given by Eqs. (2). Physically, this is because the photons in the ideal state have zero uncertainty in their relative momenta, and by the Heisenberg uncertainty principle, the photons must have infinite uncertainty in their relative positions, meaning that the photons *never* arrive at the same place together to produce a multiphoton absorption event. In general, a multiphoton state that enhances the lithographic resolution must have reduced uncertainty in the relative momenta of the photons in order to obey the resolution limit, leading to increased uncertainty in the relative positions and a reduced probability of all photons arriving at the same place.

In conclusion, while entangled photons can have a higher accuracy in where they arrive together, they are constrained by the resolution limit to arrive together less often, so there is a trade-off between feature size reduction and absorption rate.

1. References

[1] A. N. Boto, P. Kok, D. S. Abrams, S. L. Braunstein, C. P. Williams, and J. P. Dowling, "Quantum interferometric optical lithography: Exploiting entanglement to beat the diffraction limit," Phys. Rev. Lett. **85**, 2733 (2000).