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Optical Projection Lithography at Half the Rayleigh Resolution Limit by Two Photon Exposure

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In recent years, with the advent of femtosecond pulse technology, two-photon absorption has commenced to be used for exposing photo-resists. It is natural to ask then, what is the spatial resolution of two-photon lithography? There has already been some discussion of resolution limits in two-photon, scanning confocal fluorescence microscopy. We will find that ordinary two-photon exposure of photo-resist merely enhances the photographic contrast, or gamma. While this improves the spatial resolution somewhat, it does so at the expense of a requirement for tighter control over the incident light intensity. Instead, we introduce a new type of exposure system employing a multiplicity of 2-photon excitation frequencies which interfere with one another to produce a super-resolution stationary image, exhibiting a true doubling of the spatial resolution.

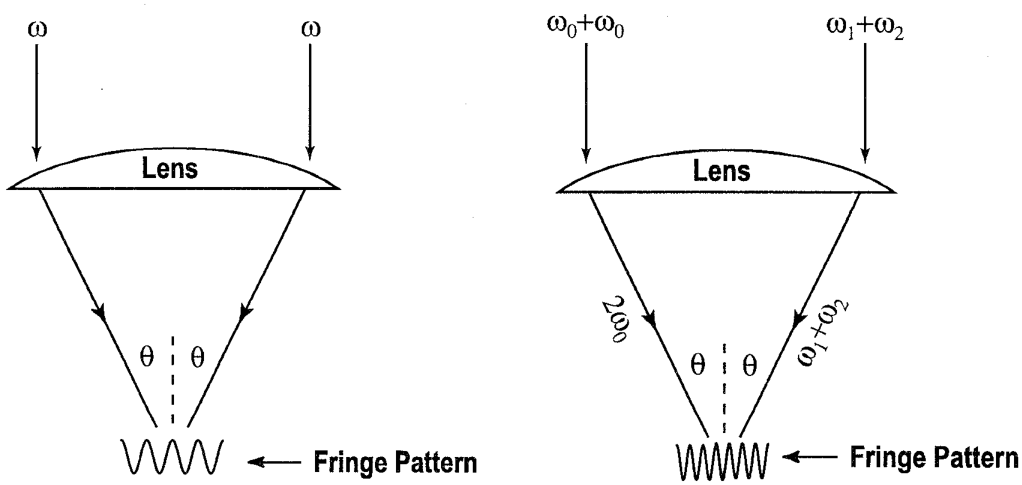

Figure 1(a): The intensity fringe pattern produced by rays converging from opposite edges of a lens. (b) The fringe pattern produced by two-photon excitation of a photoresist, in which the incident rays on opposite sides of the lens are separated into distinct frequency groupings. The frequency differences cause normal resolution fringes to oscillate rapidly in time, and they get washed out.

We can examine the resolution limits by studying the fringe pattern produced by two rays converging from opposite edges of a lens as illustrated in Figure 1(a). Such a fringe pattern is an indication of the resolution limits. Figure 2(a) shows the intensity fringe pattern in the focal plane, that results from two rays of individual wave vector k, converging at an angle θ. The spatial period of the fringes is determined by wave-vector of magnitude $K = 2k\sin\theta$. The distribution of intensity, I(x), in the focal plane is simply $I(x) = (1+\cos Kx)$.

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Since two-photon absorption is proportional to the square of intensity, the two-photon exposure will be proportional to:

$$(1+\cos Kx)^2 = \left(\frac{3}{2} + 2\cos Kx + \frac{1}{2}\cos 2Kx\right), \qquad \text{.........(1)}$$

a functional dependence which is plotted in Fig. 2(b). We can see that the fringe pattern of two-photon absorption in Eq'n. (1) and Fig. 2(b), is a mixture of a normal resolution image represented by the cos Kx term and a super-resolution image represented by cos $2Kx$. The super-resolution image at cos $2Kx$ consists of spatial harmonics at the wave-vector $2K$, and would represent a doubling of the spatial resolution over ordinary one-photon lithography. Unfortunately the image is a mixture of both resolutions. Our goal in this paper is to show how to enhance the super-resolution image, and to suppress the normal resolution image, to the greatest degree possible.

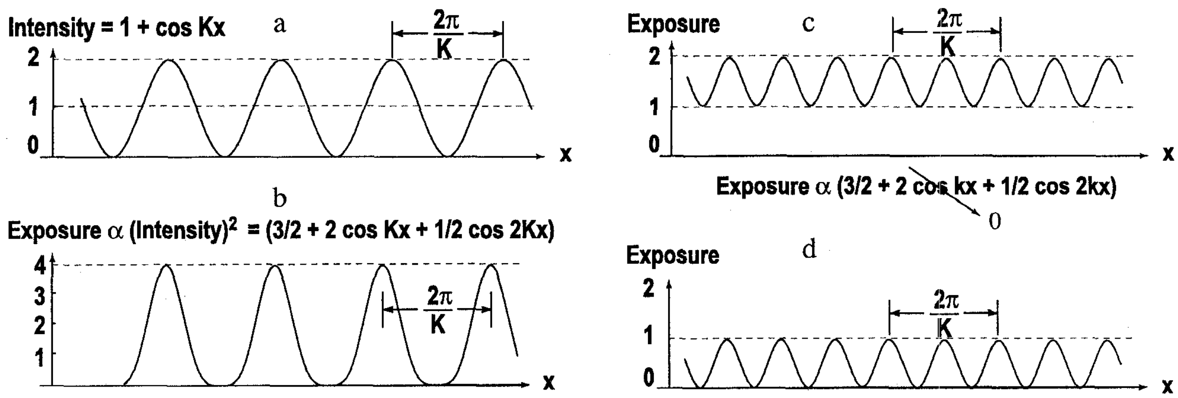
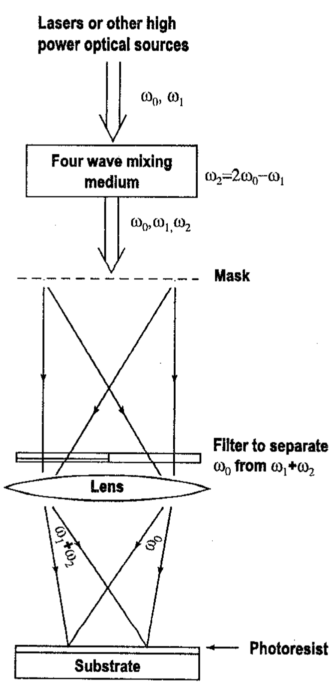


Figure 2(a): The ordinary intensity fringe pattern produced by converging rays as in Fig. 1(a), where $K = 2k\sin\theta$. (b) The intensity squared fringe pattern which consists of a normal resolution spatial harmonics at K, a super-resolution spatial harmonic at $2K$, as well as a constant term. (c) The effect of canceling the normal resolution spatial harmonic at K. The super resolution component at $2K$ remains, on a constant background. (d) By employing a photoresist with a sharp atomic transition at $2\hbar\omega_o$, the constant background is also eliminated, leaving a pure super-resolution image.

The method, or trick, for eliminating the normal resolution image is indicated in Fig. 1(b). The idea is to destroy stationary interference patterns corresponding to the linear image, while maintaining a stationary interference pattern for the super-resolution image. Arrange the incident rays to consist of ω_o on one edge of the lens, and two other frequencies slightly above and below ω_o: i.e. $\omega_1 = \omega_o+\delta$, and $\omega_2 = \omega_o-\delta$, on the other edge of the lens, where $2\omega_o=\omega_1+\omega_2$. Fringes resulting from the interference of ω_o and ω_1 oscillate rapidly at the difference frequency δ, and they wash away, averaging to a uniform background. Thus the normal resolution image goes away. Since the frequencies $2\omega_o$ and $\omega_1+\omega_2$ are coherently related, derived from four-wave mixing $\omega_2=2\omega_o-\omega_1$ for example, the super-resolution fringes are stationary and they don't wash away. This produces the result indicated in Fig. 2(c). If desired, the smeared out background on which the super-resolution image is superimposed, can be entirely eliminated. This can be accomplished by arranging for a relatively sharp atomic transition at $2\hbar\omega_o$, inside the photoresist. Then the background two-photon transitions at $\omega_0+\omega_1$, $\omega_0+\omega_2$, $2\omega_1$, and $2\omega_2$ in Fig. 1(b) do not occur, and the smeared out background becomes absent. This results in the pure super-resolution fringe pattern of Fig. 2(d) at a spatial wave vector of $2\vec{K}$ with no background, and is the ultimate culmination of two-photon lithography.

A possible exposure system is shown in Fig. 3. Very short pulses of high peak power are needed to produce a damage-free two-photon exposure in the photoresist. Fortunately such pulses can be readily frequency converted to provide new optical frequencies, for example by four wave mixing as shown in Fig. 3. The various frequencies can illuminate a mask, even a phase mask, as in conventional photo-lithography, but near the lens the various frequencies become spatially separated by spectral filters. The lens can be divided into zones, some of which pass certain frequency combinations, and some which pass other combinations. For example in Fig. 3 the lens is divided into two zones. The frequency ω_0 passes through one zone and the frequencies ω_1 and ω_2 pass through the other zone. Miscellaneous variations on the optical layout in Fig. 3 are possible, including the insertion of optical gain, and various ways of generating ω_0, ω_1 and ω_2, etc. Fig. 3 also displays various numerically computed images of a narrow, sharp, spatial line source, as limited by the finite numerical aperture (N. A.) of a lens, treating the problem one-dimensionally. We take in this case, N. A.=0.5, expressing the image size in units of the fundamental wavelength $\lambda \equiv 2\pi c/\omega_0$. A description of each curve is given in the figure caption. For 2-photon exposure with 2 zones, the resolution has already almost doubled. More zones give further improvement.



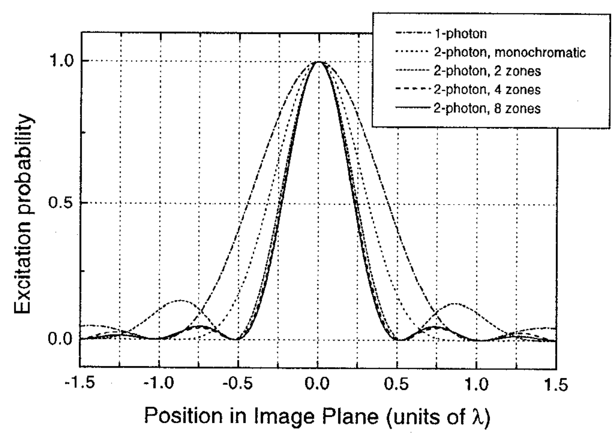


Figure 3: Left: Layout of a two-photon exposure system. Right: Computer-calculated image of a line source at infinity. The dash-dotted line represents the intensity distribution at the focus of a lens by linear diffraction through a numerical aperture=0.5. The dotted line is the intensity squared distribution associated with two photon absorption from a monochromatic beam. The other three curves represent multi-frequency two-photon excitation in a photoresist with a two, four and eight zones respectively. Whether two or eight zones the resolution is essentially twice as good as for the dash-dotted ordinary one-photon exposure.