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Defocus as an ineffective means of changing spot size for fluctuation microscopy

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Abstract. We examine the use of defocus for changing the effective spot size in fluctuation microscopy. Our conclusions relate to other coherent techniques. While it is possible to increase the illuminated spot size by defocusing, the number of phase oscillations inside the illuminated spot increases. As a result the stationary phase region increases with defocus at a slower rate than the spot size. Therefore, the effective spot size for fluctuation microscopy is not the beam size, and the phase interference is increasingly diluted by incoherent contributions, rendering defocus an ineffective means of changing the spot size for fluctuation microscopy.

Fluctuation microscopy (FM) [4, 5] examines medium-range order in disordered materials. The method relies on the measurement of statistical fluctuations in the diffraction by a coherent probe from an ensemble of sample volumes, and can be implemented using coherent nanodiffraction patterns obtained in a scanning transmission microscope. Modifying the illuminated spot size is key to the technique. The method has been used with light [6] and x-rays [7], although electrons have most widely been used[4]. The fluctuations, often measured simply by examining the variance of the scattering with $Q$, have been shown to be very sensitive to medium range order [4]. A particularly simple way to measure a correlation length has been proposed in which the illuminated probe size is varied and the correlation length plotted as a function of the probe size [8].

1. Theory for a Gaussian probe

The conditions for FM require a coherently illuminated region centered over an object. The object is usually much larger than the illuminated region, and data is recorded by moving the object and statistically analyzing the fluctuations in the diffraction patterns. To simplify our discussion we assume that the illumination wavefunction intensity is a Gaussian with standard deviation $\sigma$, with a constant phase in the illumination plane where the thin object sits. The amplitude is $\psi = \psi_0 \exp(-r^2/4\sigma^2)$ where the radial coordinate $r$ is relative to the beam center. Within the Fresnel approximation, defocusing is equivalent to replacing the standard deviation with a
complex $\sigma' = \sqrt{\sigma^2 - i\lambda \Delta f/4\pi}$ where $\lambda$ is the radiation wavelength and $\Delta f$ the defocus. The resulting amplitude of the illumination is still a Gaussian, with an increased standard deviation $\sigma' = \sigma \sqrt{1 + \Delta^2}$, where the “generalized” defocus is $\Delta = \lambda \Delta f/4\pi \sigma^2$. The phase distribution in the object plane in the reduced coordinate $u = r/\sigma$ is given by

$$
\phi(u) = \tan^{-1}(\Delta) - \frac{\Delta u^2}{2(1 + \Delta^2)}
$$

(1)

The parabolic phase distribution in equation (1) is not desirable for FM, because the theory requires that the phase be constant (or linearly varying) across the illumination [5]. Since the technique works by summing the scattered components from the illuminated atoms and examining the interference between them, the change in phase across the illumination scrambles the interference. Phase oscillations within the Gaussian envelope are demonstrated in fig. 1 for $\Delta=2$.

There are two serious problems caused by this oscillatory phase: the region over which coherent interference could occur as though the phase was stationary is limited to a smaller region in the center of the probe, and the speckle amplitude is diluted by the “incoherent” scattering from the oscillatory regions. We define the “stationary” phase region as a circular region of radius $u_c$ at the center of the Gaussian where the phase change is limited to less than $\pi/4$. (Note that the absolute value of the phase at the origin, $\tan^{-1}(\Delta)$, does not affect any measurements). From equation (1)

$$
u_c = \sqrt{\frac{\pi(1 + \Delta^2)}{2\Delta}}
$$

(2)

The critical radius $u_c$ grows with $\Delta$ but more slowly than the intensity standard deviation does. Figure 2 shows the functional forms of $u_c$ and $\sigma/\sigma$. Asymptotically the critical radius goes like $\sqrt{\Delta}$ whereas the amplitude half-width grows like $\Delta$.

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**Figure 1**: the amplitude (dotted) and phase (solid) distribution of a defocused Gaussian beam (with $\Delta=2$) the phase oscillations represent a shift of $\pi$, and the amplitude is arbitrary.

**Figure 2**: The standard deviation for the amplitude (dashed) and the critical radius of the stationary phase region (solid) as a function of generalized defocus for a Gaussian beam.

**Figure 3**: The integrated contributions with increasing distance $u$ to the coherent signal from a chain of correlated atoms at $\Delta=2$, where $u_c=1.99$.

**Figure 4**: The coherent fraction of the illumination as a function of the generalized defocus.
Fig. 3 shows that only the region up to about the critical radius contributes to the coherent speckle from an ordered region, and that the oscillatory contributions beyond average to zero. So the maximum correlation length which is being probed is \( u_c \) and not \( \sigma_d \).

The situation is made worse by the fact that the oscillatory parts of the phase add to the incoherent signal, which reduces the signal to noise of the coherent signal that is sought in FM. The coherent fraction can be estimated by integrating up to \( u_c \) and normalizing to the total integrated intensity to give the function shown in figure 4

\[
C(\Delta) = 1 - \exp\left(-\frac{\pi \Delta}{8}\right)
\]  

(3)

So now we can see the problem. For example, to get the effective coherent illumination size to increase to \( r = 2 \sigma \), it is necessary to use a generalized defocus of 2.06 where the coherent fraction is only 17%. Attempts to usefully increase the critical radius above \( \sigma \) are thus severely hampered by the reduced visibility of fringes, and the actual coherent spot size is NOT the same as the defocused spot size (but can in principle be calculated from it). Since FM often relies on the height of speckle peaks as well as just their existence [5], this systematic change in visibility must be at very least accounted for, and most likely renders the signal-to-noise too low for useful application.

While these results were calculated specifically for Gaussian illumination, we expect them to apply to other illumination functions, since the parabolic nature of the emerging wave front, while perturbed by the detailed amplitude distribution, is general, especially at large defocus \( \Delta >> 1 \).

2. Comparison to experiment and discussion

Despite the theory above, there appears to be experimental evidence that the coherent illuminated region can be varied by defocusing, both for x-rays [2, 9] and for electrons [3]. However, we can explain this paradox by the simple assumption that the structures that were examined were dominated by random correlations, which is often the case. With random structures, the existence of the phase curvature in the wavefront is just equivalent to an additional random phase factor. In other words no matter what the phase structure for the probe, provided it is well-defined, there will be a combination of scatterers in a random structure which will create speckle coming from atoms across the entire illuminated region. Just by observing the reciprocal space size of speckle from a random sample, one cannot identify the presence of phase curvature. On the other hand, in FM, where you are looking for a signal from medium-range ordering, the phase curvature will scramble the correlations outside the critical radius as described above. While it may be possible to account for this effect and still use the technique, on simple signal-to-noise grounds we conclude that the defocusing technique is not effective for FM. Phase curvature is also likely to affect many other applications of coherent diffraction, such as Coherent Diffraction Imaging which we have considered elsewhere [2].

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