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RETRIEVING OF THE COMPLEX DEGREE OF SPATIAL COHERENCE OF ELECTRON BEAMS

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Abstract

We discuss the applicability of a recently developed method for two-dimensionally retrieving the complex degree of spatial coherence of laser beams, in both amplitude and phase, to the case of the electron beam provided by the source of an electron microscope. Obtaining an electron beam with the highest possible coherence is critical for successful holography experiments. Therefore, the accurate measurement of the complex degree of spatial coherence is highly desirable. The method consists of the following three steps: recording of the beam spot, determining its centered-reduced moments and inserting them as coefficients of a series. This procedure is simple, fast and of higher performance than conventional procedures such Fourier analysis or Young interferometry. Experimental results are presented.

1. FUNDAMENTALS

The complex degree of spatial coherence is well defined in optics [1, 2] for describing the correlation between the optical field fluctuations at two different points of a specific plane. Its modulus takes values in the interval [0,1], with zero for uncorrelated fluctuations and one if their correlation is maximal.

On the other hand, the concept of *coherence* is also applied to particles, which move along indistinguishable paths and give rise the interference patterns, understanding *indistinguishability* in the sense of the quantum mechanics [3, 4]. In this case, the wave functions of the particles will be correlated in some extent, and a complex degree of spatial coherence can be defined for describing this feature. Indistinguishability is also established when two wave packets associated with two particles are not resolvable, namely, the wave packets are superimposed and we cannot follow the particles in specific and differentiable trajectories [3]. Thus, coherence properties allow the establishment of the fundamental link between the wave and the particle descriptions [4].

Correlation of optical field fluctuations and of wave functions of the particles determines the capability of light and particle beams, respectively, of producing interference patterns, in such a way that the modulus of the complex degree of spatial coherence will be related to the visibility of the interference fringes, and its phase to shifts of the fringes with respect to the co-ordinate origin. However, it is possible to retrieve the complex degree of spatial coherence of optical fields, in both amplitude and phase, by applying a non-interferometric procedure [5-7], i.e by using the centered-reduced moments of the spot. In this paper, we discuss the applicability of this procedure for determining the complex degree of spatial coherence of electron beams in field-free domain.

If the electrons of the beam are completely indistinguishable, their propagation is described by the solution of the Schrödinger equation independent of time [8]. This equation takes the form of the Helmholtz equation

$$\nabla^2 \Psi(\mathbf{r}) + k^2 \Psi(\mathbf{r}) = 0\,, \tag{1}$$

where $\Psi(\mathbf{r})$ is the wave function for electrons, which depend on the position vector **r**, and $k = \frac{g}{\hbar} = \frac{2\pi}{\lambda}$, g being the constant kinetic momentum and λ the corresponding De Broglie's wavelength. Following the optical equivalent [1], the solution of equation (1) should fulfil the Kirchhoff's general diffraction formula. By regarding the electron beam in paraxial approach, this formula takes the form used for describing Fraunhofer diffraction, i.e.

$$\Psi(x', y') = -\frac{i}{\lambda z} e^{ikz} \iint_A \Psi_0(x, y) e^{-i\frac{k}{z}(x'x + y'y)} dx\, dy\,, \tag{2}$$

with (*x,y*) and (*x',y'*) the Cartesian co-ordinates on planes orthogonal to the direction of propagation *z*. This wave function should be of deterministic nature.

However, from a more realistic point of view, the wave function $\Psi(\mathbf{r}, t)$ should be a random variable that represents a fluctuating electron beam in space-time $(\mathbf{r}, t)$. So, the behaviour of the beam can be more appropriately described by the correlation of the wave function at two points of the space-time $(\mathbf{r}_1, t_1)$ and $(\mathbf{r}_2, t_2)$, i.e.

$$\Gamma(\mathbf{r}_1, \mathbf{r}_2, t_1, t_2) = \left\langle \Psi^*(\mathbf{r}_1, t_1)\, \Psi(\mathbf{r}_2, t_2) \right\rangle, \tag{3}$$

where the angular parenthesis represents ensemble average, in the sense of the second-order spatial coherence theory [2, 8]. Equation (3) indicates that the electron beam should be a stochastic process. In field-free domain, it can be considered as stationary and ergodic, at least in the wide sense [2, 8], so that equation (3) only depends explicitly on the time difference $\tau = t_1 - t_2$. Thus, it becomes

$$\Gamma(\mathbf{r}_1, \mathbf{r}_2, \tau) = \left\langle \Psi^*(\mathbf{r}_1, t)\, \Psi(\mathbf{r}_2, t - \tau) \right\rangle. \tag{4}$$

Following the same reasoning applied in Refs. [1, 2] for the mutual coherence function of stationary and ergodic (at least in a wide sense) optical fields, we conclude that the function $\Gamma(\mathbf{r}_1, \mathbf{r}_2, \tau)$ should satisfy two coupled Schrödinger equations of the form

$$-\frac{\hbar^2}{2m} \nabla_j^{\,2} \Gamma(\mathbf{r}_1, \mathbf{r}_2, \tau) = -i\hbar \frac{\partial}{\partial \tau} \Gamma(\mathbf{r}_1, \mathbf{r}_2, \tau), \tag{5}$$

where m denotes the (non relativistic) electron mass, j=1, 2 specifies the co-ordinates $\mathbf{r}_j$ and $i = \sqrt{-1}$. On the other hand, because of the properties attributed to $\Gamma(\mathbf{r}_1, \mathbf{r}_2, \tau)$, it is possible to postulate the existence of its Fourier spectrum $W(\mathbf{r}_1, \mathbf{r}_2; \nu) = \int_{-\infty}^{\infty} \Gamma(\mathbf{r}_1, \mathbf{r}_2; \tau) e^{2\pi i \nu \tau}\, d\tau$, with ν the frequency of the electron wave, which can be expressed as the spectral correlation [2, 8]

$$W(\mathbf{r}_1, \mathbf{r}_2; \nu)\, \delta(\nu - \nu') = \left\langle \tilde{\Psi}^*(\mathbf{r}_1, \nu)\, \tilde{\Psi}(\mathbf{r}_2, \nu') \right\rangle, \tag{6}$$

with $\tilde{\Psi}(\mathbf{r}, \nu)$ the Fourier spectrum of $\Psi(\mathbf{r}, t)$ and $\delta(\nu - \nu')$ the Dirac delta [9]. The spectral correlation $W(\mathbf{r}_1, \mathbf{r}_2; \nu)$ should satisfy two coupled Helmoltz equations, as obtained by Fourier transforming equation (5), that is

$$\nabla_j^{\,2} W(\mathbf{r}_1, \mathbf{r}_2; \nu) + \frac{4m\pi\nu}{\hbar} W(\mathbf{r}_1, \mathbf{r}_2; \nu) = 0. \tag{7}$$

It means that the spectral correlation constitutes a (second order) wavefront, which describes the correlated fluctuations of the electron beam in space. Thus, the solution of equation (7), for an electron beam in paraxial approach, will satisfy a second order diffraction formula, based on the Fraunhofer diffraction integral in equation (2). This formula yields the evolution of the beam fluctuations along its propagation from a plane P1 to a plane P2, placed at a distance z to each other in Fraunhofer domain, i.e.

$$W_{P2}\left(x_A + \frac{x_D}{2}, x_A - \frac{x_D}{2}, y_A + \frac{y_D}{2}, y_A - \frac{y_D}{2}; \nu \right) = \frac{m\nu}{\pi \hbar z^2}$$

$$\iiiint W_{P1}\left(\eta_A + \frac{\eta_D}{2}, \eta_A - \frac{\eta_D}{2}, \xi_A + \frac{\xi_D}{2}, \xi_A - \frac{\xi_D}{2}; \nu \right) e^{-i\frac{k}{z}(\eta_A x_D + \eta_D x_A + \xi_A y_D + \xi_D y_A)} d\eta_A\, d\eta_D\, d\xi_A\, d\xi_D \tag{8}$$

where $k = \sqrt{\frac{4m\pi\nu}{\hbar}}$. The pairs of positions $(\mathbf{r}_1, \mathbf{r}_2)_{P1}$ and $(\mathbf{r}_1, \mathbf{r}_2)_{P2}$ were expressed in terms of the same centre and difference co-ordinates used in Refs. [6, 7], i.e. $(\eta_A, \eta_D; \xi_A, \xi_D)$ and $(x_A, x_D; y_A, y_D)$ respectively. Replacing the De Broglie's relation for k in the former expression yields the Einstein's relation between the oscillating frequency and the total energy of the free electron, $E = \hbar\omega$ with $\omega = 2\pi\nu$.

Now, on account of the Schwartz inequality [9] equation (6) becomes

$$W(\mathbf{r}_1, \mathbf{r}_2; \nu) = \mu(\mathbf{r}_1, \mathbf{r}_2; \nu) \left\langle \left| \tilde{\Psi}(\mathbf{r}_1, \nu) \right|^2 \right\rangle^{\frac{1}{2}} \left\langle \left| \tilde{\Psi}(\mathbf{r}_2, \nu) \right|^2 \right\rangle^{\frac{1}{2}}, \tag{9}$$

where $\mu(\mathbf{r}_1, \mathbf{r}_2; \nu) = |\mu(\mathbf{r}_1, \mathbf{r}_2; \nu)| e^{i\alpha(\mathbf{r}_1, \mathbf{r}_2; \nu)}$ is a hermitian quantity, i.e. $\mu(\mathbf{r}_1, \mathbf{r}_2; \nu) = \mu^*(\mathbf{r}_2, \mathbf{r}_1; \nu)$ with the asterisk denoted complex conjugated, that fulfils the conditions $0 \le |\mu(\mathbf{r}_1, \mathbf{r}_2; \nu)| \le 1$, $|\mu(\mathbf{r}, \mathbf{r}; \nu)| = 1$ and $\alpha(\mathbf{r}, \mathbf{r}; \nu) = 0$. This quantity is a measurement of the degree of correlation between the beam fluctuations at two points on a specific plane, i.e. a measurement of the indistinguishability of the electrons in the beam. Following the optical analogy, we call it the *complex degree of spatial coherence* of the electron beam.

In cases of practical interest, such as electron microscopes, the beam can be prepared with the following properties: $\mu_{P1}(\mathbf{r}_1, \mathbf{r}_2; \nu) = \mu_{P1}(\mathbf{r}_1 - \mathbf{r}_2; \nu)$ and $\left\langle \left| \tilde{\Psi}_{P1}(\mathbf{r}_1, \nu) \right|^2 \right\rangle^{\frac{1}{2}} = \left\langle \left| \tilde{\Psi}_{P1}(\mathbf{r}_2, \nu) \right|^2 \right\rangle^{\frac{1}{2}} = \left\langle \left| \tilde{\Psi}_{P1}(\nu) \right|^2 \right\rangle^{\frac{1}{2}}$. Consequently, equations (8) and (9) yield

$$W_{P2}\left(x_A + \frac{x_D}{2}, x_A - \frac{x_D}{2}, y_A + \frac{y_D}{2}, y_A - \frac{y_D}{2}; \nu \right) = \frac{m\nu}{\pi \hbar z^2} \left\langle \left| \tilde{\Psi}_{P1}(\nu) \right|^2 \right\rangle \iint_{P1} e^{-i\frac{k}{z}(\eta_A x_D + \xi_A y_D)} d\eta_A\, d\xi_A$$

$$\iint_{P1} \mu_{P1}(\eta_D, \xi_D; \nu) e^{-i\frac{k}{z}(\eta_D x_A + \xi_D y_A)} d\eta_D\, d\xi_D \tag{10}$$

for this kind of beams. A detector attached at the plane $P2$ will record the quantity $S_{P2}(x_A, y_A; \nu) = W_{P2}(x_A, x_A, y_A, y_A; \nu)$ which is proportional to the probability to find an electron in a given region (which is associated with the "intensity" of the beam [8].) i.e., it is related to the mean value of the square modulus of the wave function. Then, from equation (10) follows

$$\frac{S_{P2}(x_A, y_A\, ;\nu)}{A_{P1}\left\langle\left|\tilde{\Psi}_{P1}(\nu)\right|^2\right\rangle} = \frac{m\nu}{\pi\,\hbar\,z^2}\iint_{P1}\mu_{P1}(\eta_D\,,\xi_D\,;\nu)\,e^{-i\frac{k}{z}(\eta_D\,x_A + \xi_D\,y_A)}\,d\eta_D\,d\xi_D\,, \tag{11}$$

with $A_{P1} = \iint_{P1} d\eta_A\, d\xi_A$. Note that the quantity $\rho_{P2}(x_A, y_A\,;\nu) = \frac{S_{P2}(x_A, y_A\,;\nu)}{A_{P1}\left\langle\left|\tilde{\Psi}_{P1}(\nu)\right|^2\right\rangle}$ satisfies the axioms of the probability density functions [9]. Consequently, it is a positive-definite function whose support fits the smallest region of sides 2A and 2B along the axes x_A and y_A at the detector plane, outside of that the values of $\rho_{P2}(x_A, y_A\,;\nu)$ are negligible. Equations (11) means that $\rho_{P2}(x_A, y_A\,;\nu)$ and $\mu_{P1}(\eta_D\,,\xi_D\,;\nu)$ constitute a Fourier pair if the integration regions over the planes *P*1 and *P*2 are big enough, i.e. significantly bigger than the De Broglie wavelength, so that equation (11) should be regarded as the Fourier transform of the complex degree of spatial coherence of the beam and the approach

$$\delta(\eta_D - \eta'_D)\,\delta(\xi_D - \xi'_D) = \frac{1}{z}\sqrt{\frac{m\,\nu}{\pi\,\hbar}}\iint_{P2} e^{-i\frac{k}{z}[(\eta_D-\eta'_D)\,x_A + (\xi_D-\xi'_D)\,y_A]}\,dx_A\,dy_A$$

holds. Under these conditions, Fourier transforming equations (11) yields

$$\mu_{P1}(\eta_D\,,\xi_D\,;\nu) = z\sqrt{\frac{\pi\,\hbar}{m\,\nu}}\iint_{P2}\rho_{P2}(x_A, y_A\,;\nu)\,e^{i\frac{k}{z}(\eta'_D\,x_A + \xi'_D\,y_A)}\,dx_A\,dy_A\,. \tag{12}$$

Equation (12) means that the complex degree of spatial coherence of the electron beam can be determined if the quantity $\rho_{P2}(x_A, y_A\,;\nu)$ is properly recorded by the detector attached at the plane *P*2. According to the optical analogy [5-7], we call it the *spot of the electron beam*.

2. RETRIEVING PROCEDURE

Following equation (12), the complex degree of spatial coherence of the electron beam should be straightforwardly recovered by Fourier transforming its spot. However, it is a procedure of low performance on account of usual experimental and technical features, such as detector inhomogenities and fluctuating response [7]. Furthermore, it is essentially a low-resolution procedure, because of the feature that its output is a set of readings of the same extension as the input. Thus, its measuring capability is severely limited.

Alternatively, a very accurate retrieving of the complex degree of spatial coherence of laser beams was recently obtained, by using the centered-reduced moments of the beam spot [5-7], which are defined as

$$\gamma_{mn} = \int_{-1}^{1}\int_{-1}^{1} x^m \ y^n \ \rho_{P2}(x,y)\, dx\, dy \ , \tag{13}$$

where $x = \dfrac{x_A}{A}$, $y = \dfrac{y_A}{B}$ are reduced coordinates and m, n=0, 1, 2, 3... Therefore, equations (12) and (13) yield

$$\left. \frac{\partial^{m+n} \mu_{P1}(\eta_D \, , \xi_D \, ; \nu)}{\partial \eta_D^m \ \partial \xi_D^n} \right|_{\eta_D = \xi_D = 0} = z \sqrt{\frac{\pi \hbar}{m \nu}} \left(i \, \frac{k}{z} \right)^{m+n} A^{m+1} \ B^{n+1} \ \gamma_{mn} \, . \tag{14}$$

So, the complex degree of spatial coherence of the beam can be expressed in terms of a Taylor series, whose coefficients are essentially determined by the centered-reduced moments of the beam spot, that is

$$\begin{aligned} \mu_{P1}(\eta_D \, , \xi_D \, ; \nu) &= \sum_{m=0}^{\infty}\sum_{n=0}^{\infty} \frac{\eta_D^m \ \xi_D^n}{m! \, n!} \left. \frac{\partial^{m+n} \ \mu_{P1}(\eta_D \, , \xi_D \, ; \nu)}{\partial^m \eta_D \ \partial^n \xi_D} \right|_{\eta_D = \xi_D = 0} \\ &= \sum_{m=0}^{\infty}\sum_{n=0}^{\infty} z \sqrt{\frac{\pi \hbar}{m \nu}} \left(i \, \frac{k}{z} \right)^{m+n} \frac{A^{m+1} \ B^{n+1}}{m! \, n!} \gamma_{mn} \ \eta_D^m \ \xi_D^n \end{aligned} \, . \tag{15}$$

This procedure is more stable against detector features because the readings will be averaged to determine each coefficient of the series. Furthermore, its resolution is much higher than that of the Fourier-based method, because its output is determined by a set of surfaces related to each input reading [5-7]. However, it is very sensitive against background noise because of the coefficients $(x_A^n \, , y_A^m)$ in each average. So, different pre-processing techniques for noise suppression should be applied for assuring the precision of the measurement, as that reported in Refs. [5-7].

3. EXPERIMENTAL RESULTS

We measured the complex degree of spatial coherence in a JEOL 2100F field-emission transmission electron microscope (TEM) for an experimental setup described below. The electron source in the JEOL 2100F is a thermoionic-assisted field-emission electron gun, and the TEM is operated at 200 kV accelerating voltage, which sets the electron wavelength at 2.5 pm. The microscope is equipped with an electron biprism in order to perform off-axis electron holography experiments. As a matter of practical importance, obtaining an electron beam with the highest possible coherence is critical for successful holography experiments, and accurate measurement of its complex degree of spatial coherence is, therefore, highly desirable.

In order to establish a basis for the measurements outlined here, we chose to investigate the coherence under conventional TEM imaging conditions rather than conditions appropriate for holography, which involve highly astigmatic illumination. Figure 1 shows a schematic of the electron-optical setup used to record data. The first condenser lens (labelled C1 in Figure 1) is used to de-magnify the patch of emission, producing a virtual source of electrons at a large effective distance from the object plane of the TEM. For our setup, C1 was fully excited, which corresponds to a virtual source with the smallest size at the farthest distance available. A 100 μm condenser aperture, positioned after C1, was used to collimate the beam.

The combination of second condenser lens and condenser mini-lens (labelled C2/CM in Figure 1) controls the size and the angle of convergence of the illumination on the sample. For our setup, C2 and CM were chosen to produce roughly parallel illumination over a very large area at the object plane of the TEM. The objective lens, (OL in Figure 1) normally used to focus the image of the object, was set to its standard operating value.

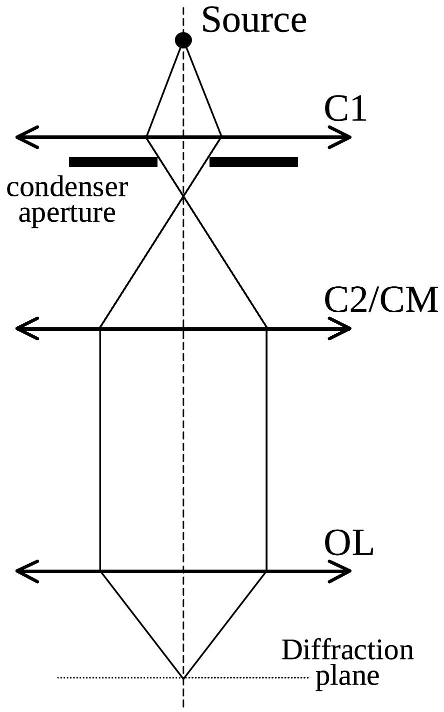
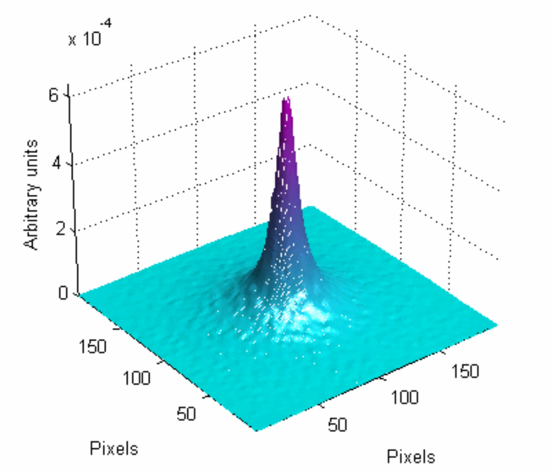
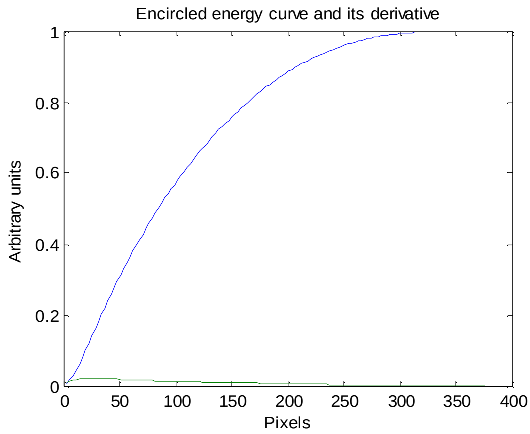


Fig. 1: Schematic of the experimental setup used to record the incident-direction diffraction spot.

The lower lenses of the microscope (not shown in Figure 1) are used to select the desired (image or diffraction) plane of the OL, and to magnify and project it onto the recording device. In this case, the diffraction plane of the OL was selected, and the incident-direction diffraction spot was recorded on a Multiscan CCD camera with a 2048x2048 array of 14 μm pixels. Calibration of the scale (in diffraction-space) of the CCD pixels was done by recording a conventional selected area diffraction pattern of gold particles dispersed on an amorphous carbon film. Due to the limited field of view provided by the CCD camera, it was impossible to directly calibrate the pixel scale for the long camera-length used in this experiment. Instead, calibration was made for a lens program corresponding to a scaled-down camera-length, and then extrapolated to the experimental settings. The camera-length was determined to be (23+/-2) m, corresponding to an exceptionally high magnification of the diffraction plane of the objective lens, and is characteristic to the specific design of this microscope. This setup is equivalent to the optical setup used for laser spot recording [7]

Figure 2a depicts the recorded electron beam spot, after the pre-processing procedures. We applied the same routines as for laser beam spots reported in Ref. [7]. To properly calculate the centered-reduced moments of the electron beam spot, the integration region for equation (13) should be determined, i.e. the region 2Ax2B. It is performed by means of the encircled energy curve [1], sketched in Figure 2b. That curve is a plot of the fraction of energy limited by a rectangle centred on the centre of mass of the beam spot, as function of its diagonal. Up to a certain diagonal length, the growth of the curve is negligible (about 300 pixels in the graph). The rectangle corresponding to this diagonal is assumed to be the integration region.





(a) **(b)**

Fig. 2: (a) Electron beam spot and (b) the corresponding encircled energy curve and it first derivative

So, 900 centered-reduced moments were calculated and inserted as coefficients in the Taylor's series of the complex degree of spatial coherence [equation (15)]. Those moments are sketched in Figure 3. Their convergence to null as the order increases is apparent. Consequently, we only need a finite number of moments to accurately retrieve the complex degree of spatial coherence.

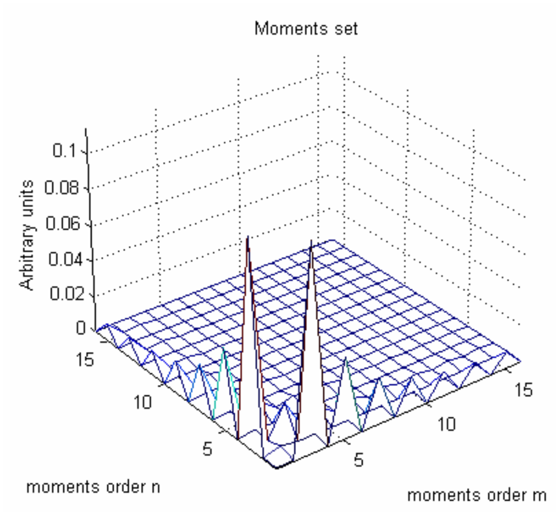
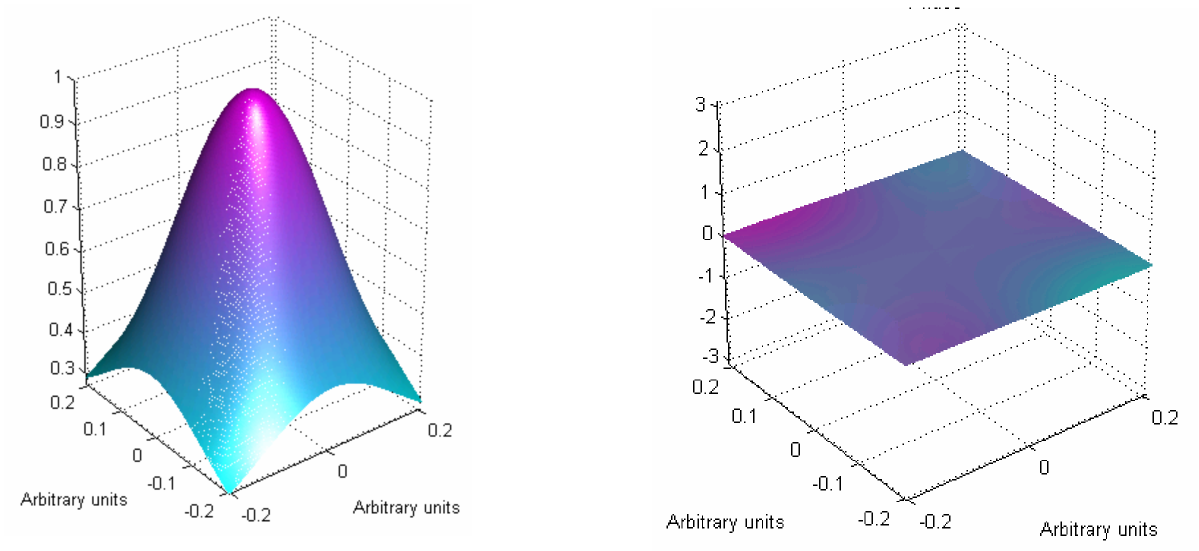


Fig. 3: Graph of the set of centered reduced moments of the electron beam spot in Figure 2. The 00-order moment was removed. Each peak is corresponding to a specific moment. The moment subscripts determine the peak position on the reticule and its value is given by the peak height. The convergence of these values to null is apparent.

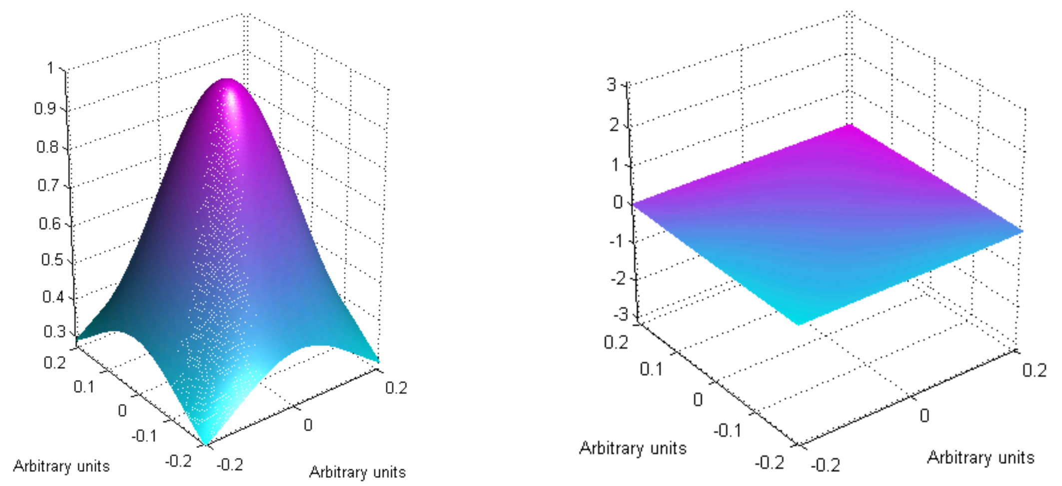
Figure 4 depicts the retrieved complex degree of spatial coherence of the spot in Figure 1, in both modulus and phase.



a. **b**

Fig. 4: Retrieved complex degree of spatial coherence of the electron beam using the centered-reduced moments procedure. a) Modulus. b) Phase.

A comparison between this method and the Fourier transform in equation (12) can be performed by regarding the results shown in Figure 5. They are the modulus and phase of the complex degree of spatial coherence of the laser beam, given by the Fourier transform of the spot in Figure 2.



a. **b.**

Fig. 5: Retrieved complex degree of spatial coherence of the electron beam using the Fourier-based method. a) Modulus and b) Phase.

It is apparent that the moduli retrieved by both methods are in agreement, but it is not the case of the phase. Instabilities in the phase retrieved by the Fourier method are mainly due to asymmetries in the recorded spot because of pixel-to-pixel variations of the individual response function, or eventual spot fluctuations during the recording [7]. The influence of these error sources on the phase retrieving by the centered-reduced moments method are significantly smaller because of the averaging procedure for moment calculation.

Consequently, the phase retrieving by the moments-based method is more accurate than that by the Fourier-based method. This feature establishes the superiority of our method.

CONCLUSION

Because of the wave nature of fluctuating electron beams, it is possible to characterize them by means of a *complex degree of spatial coherence*. Consequently, the accurate measurement of this quantity acquires a noticeable roll in electron beam characterization and quality control for applications such as electron holography. In this context, the centered-reduced moments of the beam spot provides a simple but robust technique for performing such a measurement, in both modulus and phase, whose performance is higher than that of the conventional methods based on Fourier analysis or Young interferometry.

The method consists of three steps, i.e. 1) recording of the beam spot, 2) determining its centered-reduced moments and 3) inserting them as coefficients of a series. To eliminate the influence of background noise, spot preprocessing should be applied. However, the method is very insensitive to detector features such as pixel-to-pixel variations of the individual response function or eventual spot fluctuations during the recording, because of the averaging procedure for moment calculation. This property, which is of significant importance for the phase determination, establishes the superiority of our method in comparison to the conventional techniques.

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