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Detection of Object Movement in the Optical Diffractograms of Electron Micrographs

Joachim Frank

Department of X-ray Crystallography at the Max-Planck Institut für Eiweiss und Lederforschung (Protein Research), Munich, and Department of Structure Research, Institute of Physical Chemistry, Technical University

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Abstract

[original text] **Detection of object movement in the optical diffractograms of electron micrographs.** Movements of the objects during an electron microscopic exposure cause characteristic modulations of the Fourier transform of the photographic density distribution, from which the nature of the movements can in turn be reconstructed. Modulations of this type were observed in optical diffractograms of a number of micrographs of amorphous carbon films and could be assigned to particular movements.

[starting here: translation]

1. Introduction

It is known from the work of Thon [1] that the wave aberrations prevailing during an exposure in the electron microscope can be determined from the optical diffraction pattern [of the micrograph] if a carbon film is used as object. The image of a statistical object1) essentially documents the electron microscopic parameters (voltage, wavelength, defocus, astigmatic defocus difference), though in an encoded way. The "decoding" [of this information] is carried out by taking the Fourier transform, for instance in the light-optical diffractometer.

We will show that beyond this information, the light-optical diffractogram contains additional clues regarding the behavior of the object during the exposure in the electron microscope. Specifically we will focus on the movements of the object, considered rigid during that time span. This study was prompted by [the analysis of] micrographs of carbon films which showed a strong periodic modulation.

2. General Description of [Object] Movements in Fourier Space.

We define $I(\mathbf{r})$ as the image intensity (in the following "image" for short) which hits the film during the exposure time for a resting, unchanging object. The coordinate system $\{\mathbf{r}\}$ refers to the resting film. We now assume that the image changes over time, for one due to radiation damage, and second due to the movements of the object (for instance, the thermal drift of the stage) and due to changes of the parameters describing the imaging process (for instance drift of the lens current, changes

[1] an object, in other words, in whose phase structure all spatial frequencies are distributed with equal probability ("white scatterer")]

of the astigmatism due to the buildup of contamination, etc.). If these changes occur over a time comparable to the exposure time T, we have

$$I = I(\mathbf{r}', t) \text{ with } \mathbf{r}' = \alpha(t)\,\mathbf{r}.$$

Here $\alpha(t)$ stands for a general time-dependent coordinate transformation. The recorded optical density will be proportional to (see [2])

$$S(\mathbf{r}') = \int_0^T I(\mathbf{r}', t)dt \tag{1}$$

Despite the blurring of the image that is due to the superposition of the component images the time dependence [of the process] can be inferred in simple cases. To this end we can use procedures known from both X-ray Crystallography and Statistical Optics.

We restrict ourselves to the case of translational movements of a rigid object, ignoring all other changes of the object. Let us assume that, on account of the movement of the object, the image moves by $\mathbf{q}(t)$ relative to its starting position, and as a consequence (1) is rewritten in the form

$$S(\mathbf{r}) = \int_0^T I(\mathbf{r}) \circ \delta(\mathbf{r} - \mathbf{q}(t))\, dt = T\, I(r) \circ 1/T \int_0^T \delta(\mathbf{r} - \mathbf{q}(t))dt, \tag{2}$$

using the notation $a(\mathbf{r}) \circ b(\mathbf{r}) = \int a(\mathbf{r} - \mathbf{r}')b(\mathbf{r}')\, d\mathbf{r}'$ for the convolution operation. Fourier-transformation of (2) and application of the convolution theorem yields

$$\mathcal{F}\{S(\mathbf{r})\} = T\, \mathcal{F}\{I(\mathbf{r})\} \cdot 1/T \int_0^T e^{-2\pi i \mathbf{k}\mathbf{q}(t)}\, dt = \mathcal{F}\{T\, I(\mathbf{r})\}\, M(\mathbf{k}) \tag{3}$$

[A side issue:] Instead of the optical density [of the film], the transparency $e^{-S(\mathbf{r})}$ is the quantity effectively being Fourier-transformed when the film is placed into the optical diffractometer. However, when the density differences $\Delta S(\mathbf{r})$ are sufficiently small, linearization around the average density S_0 is possible, leading to

$$e^{-S(r)} \approx (1 + \Delta S(\mathbf{r}))\, e^{-S_0} = S'(\mathbf{r}).$$

As the additive term produces only an irrelevant modification to the zero term of the Fourier transform, we will not distinguish in the following between $S(\mathbf{r})$ and $S'(\mathbf{r})$.

The intensity obtained in the diffractogram becomes therefore proportional to

$$\mathcal{F}\{S\} \cdot \mathcal{F}^*\{S\} = \mathcal{F}\{T\, I\}\, \mathcal{F}^*\{T\, I\} \cdot M(\mathbf{k})M^*(\mathbf{k}) \tag{4}$$

Now when a carbon film is used as object, the diffractogram shows a distribution approximately given by

$$|\mathcal{F}\{S\}|^2 = \sin^2 \gamma(\mathbf{k}), \quad \text{[except for a falloff term not mentioned here]}$$

where $$\gamma(\mathbf{k}) = \gamma(\theta,\varphi) = \frac{\pi}{2\lambda} C_s\, \theta^4 - \frac{\pi}{\lambda} \theta^2 [\Delta f + \tfrac{1}{2} \Delta f_A \sin 2\varphi]$$

modulated by a [two-dimensional] function MM* whose shape depends on the function $\mathbf{q}(t)$. $\mathbf{k}$ and (θ,φ) are Cartesian and polar coordinates in the plane of the

diffractogram. Further definitions are as follows:

C_s = spherical aberration constant of the objective lens
Δf = defocus
Δf_A = focus difference due to axial astigmatism
λ = wavelength of the electron

The approximation lies in the replacement of density differences by proportional differences in transparency, described above, and the assumption of a linear relationship between [scattered wave] amplitudes and intensities in the image plane of the electron microscope, which is known to hold in good approximation for bright field imaging of weak phase objects.

3. Special Movements

Without loss of essentials we can restrict ourselves to translations along a straight line. Let the image move into the x-direction,

$$\mathbf{q}(t) = \{x(t), 0\}.$$

We consider three special cases:
(a) *Jump drift* during the exposure. After $t = T_1$: the image has the position $x = 0$ until time T_1, then jumps into a new position $x = \xi$ and stays there till the end of the exposure $t = T$.

$$x(t) = \begin{cases} 0 \text{ for } 0 \le t \le T_1 \\ \xi \text{ for } T_1 \le t \le T \end{cases} \tag{5}$$

With the introduction of $\eta_1 = T_1/T\,;\ \eta_2 = T_2/T\,;\ T = T_1 + T_2\,;\ \eta_1 + \eta_2 = 1$ (6)
we obtain

$$M(\mathbf{k}) = M(k_x) = \eta_1 + (1 - \eta_2)e^{2\pi i k_x \xi} \tag{7}$$

$$MM^* = 1 + \eta_1 \eta_2 [\cos(2\pi k_x \xi) - 1] \tag{8}$$

The modulating function thus creates a pattern of equidistant stripes whose period is inversely proportional to the size of the movement, and whose amplitude depends upon the dwell times in the two positions.

We call now the ratio of periodic to constant portions of MM* the degree of modulation:

$$G = \frac{2\,\eta_1 \eta_2}{1 - 2\,\eta_1 \eta_2} \tag{9}$$

Since G can be determined from the diffractogram if – from a control experiment – the undisturbed distribution $|\mathrm{F}\{I\}|^2$ is known, the dwell times T_1 and T_2 can be determined.

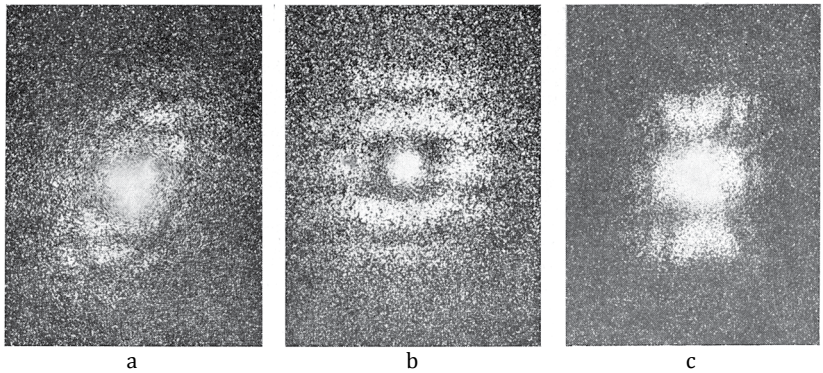
G becomes largest when $\eta_1 = \eta_2$, hence $G = 1$, and (8) now becomes

$$MM^* = \tfrac{1}{2}\,[1 + \cos(2\pi k_x \xi)] \qquad (8')$$

Fig. 1a-c depicts diffractograms of a single micrographs of a carbon film showing a modulation by $\sin^2 \gamma(\mathbf{k})$ and an additional modulation which can be explained by assuming a sudden jump (a translation) of the object during the exposure. Fig. 2 shows the electron micrograph of a contaminated graphite foil along with its diffractogram. (The contamination has the effect that the Fourier transform is dominated by a strong continuous contribution from an amorphous structural component; this is why neither the image nor the diffractogram can be distinguished from the case of an amorphous object.) One recognizes in the micrograph a preferred orientation, which runs in the direction orthogonal to the direction of the stripes in the diffractogram (Fig. 2b). Finally, Fig. 2c represents the theoretically expected intensity distribution in the diffractogram.

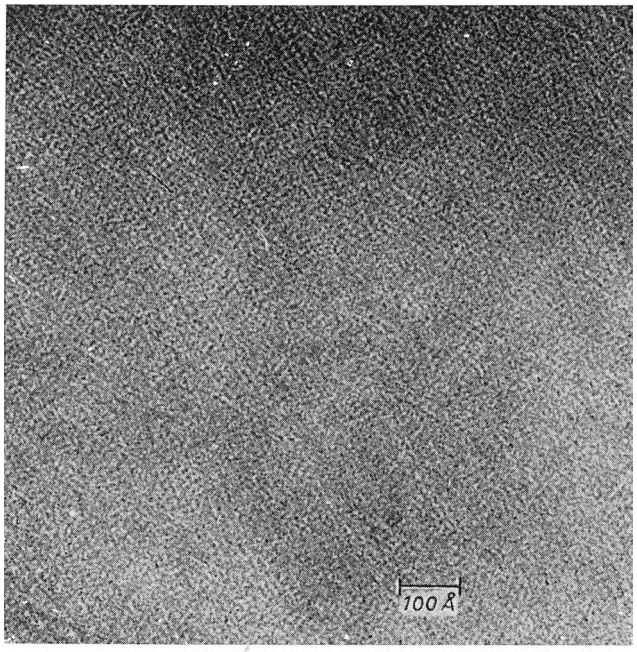
That we are not dealing with properties of the object can be demonstrated by examining other micrographs of the same object taken at different defocusses, which lack the stripes.

As mentioned at another place (5), one obtains the same periodic modulation if one takes two images of the same area, and brings these first into perfect registration in the diffractometer, then applies a small shift.

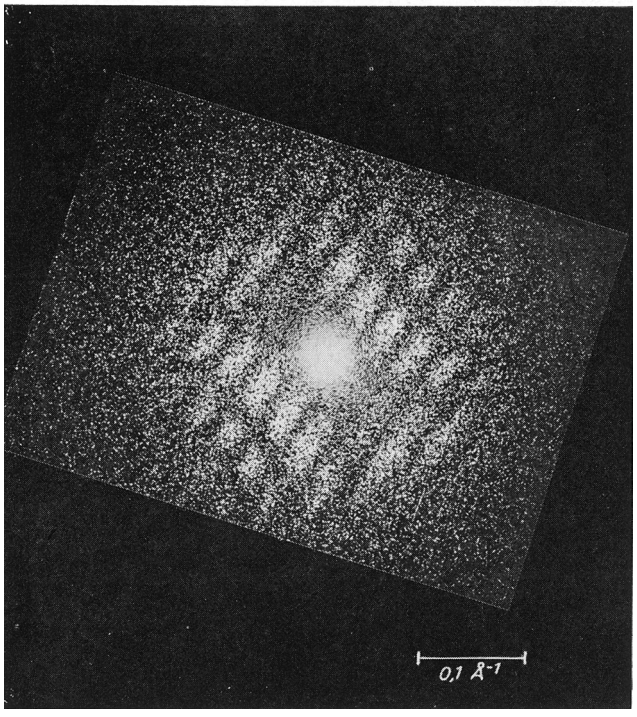


a b c

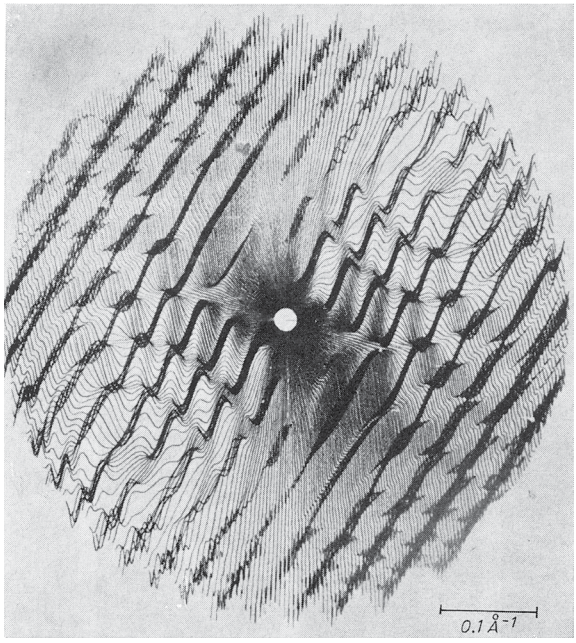
Fig. 1. Examples for the modulation of the diffractogram due to jump-drift. (U = 80kV, V_{elopt} = 220,000 x). Object: amorphous carbon film.
(a) ξ= 20 Å, Δf = 4100 Å, Δf_A = 2100 Å
(b) ξ= 17 Å, Δf = 2000 Å, Δf_A = 200 Å
(c) ξ= 7 Å, Δf = 1900 Å, Δf_A = 2100 Å



2a



2b



2c

Fig. 2. Further example for the detection of jump-drift. (U = 80kV, V_{elopt} = 160,000 x). Object: contaminated graphite film.
(a) Electron micrograph with $\Delta f \approx 1200$Å, $\Delta f_A \approx 3800$Å.
(b) Light-optical diffractogram of (a) displayed in matching orientation; $\xi = 30$Å.
(c) Representation of the expected intensity distribution using a plotter linked to an IBM 1130 computer. $MM^* \sin^2 \gamma(\mathbf{k})$ was computed using the parameters in (b), in terms of U, V_{elopt}, C_s, Δf, Δf_A, ξ, and for $\theta \leq 1.2 \times 10^{-2}$.

b) *Continuous drift:* Due to a steady drift, let the object move with constant velocity v during the exposure of length T.

$$x_D(t) = v\,t \tag{10}$$

For such a case of blurring by linear motion (see also ref. 6), some procedures for recovery of the undisturbed image have already been given (7).

We now obtain with (10)

$$M(k_x) = \frac{1}{T}\int_0^T e^{2\pi i k_x v t}\,dt = \frac{\sin(\pi k_x v T)}{\pi k_x v T}, \tag{11}$$

up to a phase factor and

$$MM^* = \left(\frac{\sin(\pi k_x \xi)}{\pi k_x \xi}\right)^2; \quad \xi = vT. \tag{12}$$

Because of the rapid decrease of the amplitudes, this distribution is not as easy to observe as in the case of (a). However, the frequent observation that the Fou-

rier plane is covered with the characteristic pattern of $\sin^2\gamma$ only within a narrow, sharply delineated stripe must be attributed to continuous drift. Fig. 3 shows a few examples for this effect, in comparison with the diffraction pattern for a normal, undisturbed exposure. The pronounced central stripe corresponds to the principal maximum of expression (12).

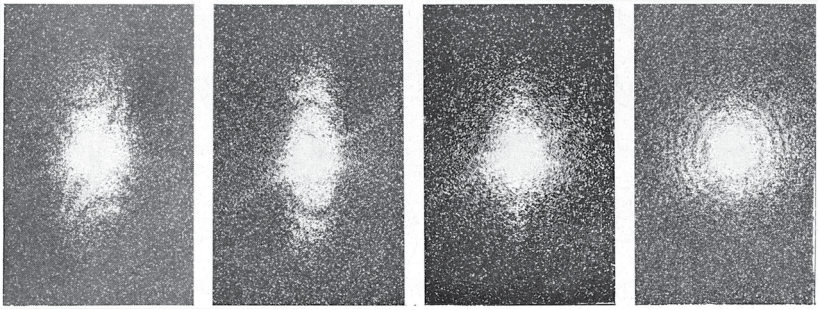


Fig. 3. Examples for modulation of the diffractogram in the case of continuous drift (U = 80 kV, V_{elopt} = 220,000 x). Object: amorphous carbon film.

(c) *Periodic oscillations:* This is another interesting case, which occurs when the object itself, or its image, oscillates relative to the photographic plate.

$$x_D(t) = \frac{\xi}{2}\cos(\omega t). \tag{13}$$

One obtains

$$M(k_x) = \frac{1}{T}\int_0^T e^{i[\pi k_x \xi]\cos(\omega t)}dt \tag{14}$$

$$= \frac{1}{T}\int_0^{wt=2\pi N} e^{i[\pi k_x \xi]\cos(\omega t)}d(\omega t) = J_0(\pi k_x \xi).$$

(Bessel function of zero order)

and

$$MM^* = J_0^2(\pi k_x \xi) \tag{15}$$

From the positions of the zeros of J_0, or the positions of the minima of J_0^2, respectively, the amplitude of the oscillations can be determined. For instance, the first zero occurs for an amplitude of ξ = 2Å at the (electron microscope's) resolution limit of 3 Å.

The term (14) also occurs in X-ray crystallography in the context of thermal vibrations. Since the amplitudes of thermal vibrations are quite small relative to the resolution achieved in electron microscopy, the term only accounts for a very small decrease of Fourier amplitudes. For an amplitude of 0.2 Å (the mean amplitude of oscillations [of an atom] at room temperature), the decrease is in the order of 1%, or 2% in quadrature.

However, oscillations that occur during the electron microscopical exposure due to mechanical vibrations of the column should be detectable for sufficiently large amplitudes.

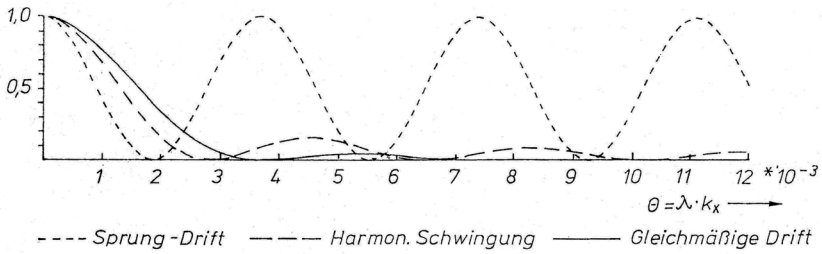


Fig. 4. Comparison of the modulation functions MM for jump-, oscillating, and continuous drift with the amplitude ξ = 10 Å.*

Figure 4 gives a comparison of modulation functions for the three cases considered, with the same amplitude parameter ξ = 10 Å. Qualitatively, the three cases can be distinguished with the help of the following criteria:
(a) *jump drift:* equidistant minima, same amplitude throughout;
(b) *continuous drift:* equidistant minima, amplitude falling off rapidly;
(c) *harmonic oscillation:* non-equidistant minima, amplitude falling off.

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References

[1] F. Thon, Z. Naturforschg. *20a* (1965) 154.
[2] R.C. Valentine, Advances in optical and Electron Microscopy I, ed.: Barer, Casslett, Academic Press, London/N.Y. 1966, 180.
[3] O. Scherzer, J. Appl. Phys. *20* (1949) 20.
[4] W. Hoppe, Acta Cryst. *10* (1957) 750.
[5] W. Hoppe, R. Langer, J. Frank, A. Feltynowski, Naturwiss. 56 (1969) 267.
[6] E.L. O'Neill, "Introduction to Statistical Optics," Addison-Wesley Publ. Comp., Reading, Mass. 1963, 27.
[7] J.L. Harris, J. Opt. Soc. Amer. *56* (1966) 569.
[8] R. Hosemann, S.N. Bagchi, "Direct Analysis of Diffraction by Matter," North-Holland Publ. Comp., Amsterdam 1962, p. 271, 286.