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On the Contents of Information from Electron Microscope Images, Especially from Defocused Series, of Crystal Lattice Periods

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As in light optics when one is faced with uncommon objects or unusual illumination, in electron microscopy of atomic structures it is hardly possible to draw conclusions upon an object structure from its image without a reasonably exact knowledge of the interaction between radiation and object. Uncertainty about this interaction, however, must be overcome to draw from the obtainable images the information of the object structure. It seems to be rather necessary to study theoretically simple cases which may help us to understand characteristic features of observations and to give suggestions for systematic experimental research in this field. With respect to ambiguities, moreover, it may become important to utilize whole defocused series. A simplified example of higher physical importance than the two-beam-case of diffraction is discussed in the following. It shows instructively which properties of the object are mainly important for the change of flux density in an electron beam as it passes through the crystal lattice, and how the initial phase contrast is gradually transformed into an amplitude contrast.

Introduction

In electron microscopy of the present time, images are often obtained from crystalline objects with a marked structural periodicity in a plane nearly normal to the direction of the incident beam. If the lattice spacing of such a periodicity is large enough to be resolved, and the diffracting power sufficiently high, the superposition of the transmitted and diffracted beams may produce a system of fringes in the microscopic image which may be interpreted as an image of the set of lattice planes which is responsible for the diffraction. The fringe structure of such an image depends not only on lattice structure, but also on angle of incidence, thickness of object, apertures of lenses and on degree of defocusing. Without a reasonably exact knowledge of the influence of these conditions, it is hardly possible to draw any conclusion, except for those concerning the presence of the basic periodicity, from the image structure.

The meaning of "Image of an Atomic Structure"

In view of the complication indicated above, we should avoid to consider the observed pattern as a true image of the structure of the object. We must of course careful about a deficient resolution which can only give a defective image. Apart from this, however, the dependence on the conditions of observation is not so different from that met in light optics when one is faced with uncommon objects or unusual illumiuation, and inadequate knowledge is available of the interactions between radiation and object. The technically faultless image of a transmitted sheet of regularly arranged balls of glass or similar non-absorbing bodies capable of refraction and reflection, allows only experienced observers to recognize the object. Without special knowledge one can deduce neither the distribution of mass nor the shape of elementary bodies, but only the basic periodicities of their geometrical array. One obtains more information about the object only if one is conversant with the changes which would occur when the illumination or other factors were varied. Image obtained with unusual illumination e.g. with infra-red radiation might not at first appear to be true ones, simply because we are not familiar with them. Apart from such custom of us, however, there is no reason for preferring the images perceived under white light as the really true ones. What, therefore, should we regard as true images of atomic structures which we can never observe with visible light? It would here be wrong to reserve the term "imaging" which is used for methods of observations other than electron microscopy, and to call electron micrographs of such structures merely interference patterns.

It cannot be doubted that the intensity distribution is generally not uniform, even at the exit surface of a non-absorbing crystalline object but will show peculiarities, e. g. fringes characteristic of the strong interaction between the electron beam and the lattice structure with respect to its thickness dimension. Thus, mostly it would be misleading to seek an interpretation of images in terms of the analogue of a plane grating. On the other hand, the image of an atomic structure certainly will never include relatively sharply bounded contours similar to those we meet in microscopy of larger objects, and images from such structures will look rather similar either in or out of focus. Then, perhaps, there would be little reason to prefer the one to the other and we should utilize whole defocused series.

The aim of the following investigation

As to such atomic structures, little is known about the change in the flux density which an electron beam suffers when it passes through the object. Multiple diffractions are nearly always to be considered. It, therefore, seems to be of interest to investigate some theoretically simple cases which may help us to understand characteristic features of observations, and to give suggestions for systematic experimental research in this field. A simplified example of somewhat higher physical importance than the two-beam-case of diffraction, which is usually treated, already revealed a remarkable dependence on both thickness and defocussing of the intensity distribution, and shows features which are probably of more general importance.

Discussion of a simplified case

It is based on the following suppositions:

I. The object has a simple sinusoidal inner potential

$$\phi(x)=\phi_0+2\phi_a\cdot\cos(2\pi x/a)\ ,\quad (\phi_a>0) \qquad (1)$$

where x is a coordinate parallel to the surface of a plate-like crystal. The planes $x=n\cdot a$ (n=integer) of maximum potential may be called the lattice planes.—II. The incidence of primary beam is normal to the surface. Then the Bragg condition is not fulfilled for the set of lattice planes, but one or more pairs of symmetrically diffracted beams may occur to either side of the planes. —III. Here, for the sake of an easy survey and simplicity, we do not use Mathieu functions as solutions, and suppose that diffraction of higher orders than ± 1 are negligible. This simplification has the advantage that it allows us to easily obtain generalizations of another kind later on.

The calculation based on the dynamical theory, gives a result represented by a formula which can easily be interpreted. The intensity distribution behind the crystal is of the form

$$|\psi|^2=1+A_1\cdot\cos(2\pi x/a)+A_2\cdot\cos(4\pi x/a) \qquad (2)$$

where the coefficients A_1 and A_2 indicate the contrast of the part of basic and half the basic periods in the fringes respectively:

$$A_1=\frac{8s\cdot\sin^2(\pi D/2D_0)}{1+8s^2}\cdot\cos(\pi z/t)+\frac{4s\cdot\sin(\pi D/D_0)}{\sqrt{1+8s^2}}\cdot\sin(\pi z/t) \qquad (3)$$

$$A_1=\frac{8s}{1+8s^2}\cdot\sin(\pi D/2D_0)\times\sqrt{1+8s^2\cos^2(\pi D/2D_0)}\cdot\sin(\beta+\pi z/t) \qquad (4)$$

$$A_2=\frac{8s^2}{1+8s^2}\cdot\sin^2(\pi D/2D_0) \qquad (5)$$

$$tg\beta=\frac{tg(\pi D/2D_0)}{\sqrt{1+8s^2}} \qquad (6)$$

Here, z is the distance of the observed point from the exit surface. Thus, the intensity in a plane z=const. describes the image obtained after overfocussing through the distance z (within the object space), and negative values of z give the virtual distributions which can be observed at underfocussing. D =thickness of the crystal, $t=a^2/\lambda$=characteristic depth of sharpness out of focus. $s=\phi_a a^2/150\ \text{VÅ}^2$ is obviously a characteristic "diffracting power" of the set of lattice planes, and $D_0=t/\sqrt{1+8s^2}$ is a characteristic thickness at which the highest possible contrast occurs.

The first term in eq. (2), the mean intensity in the fringe system, equals the uniform intensity (taken as unity) of the primary beam. The second term describes the expected basic periodicity of the object. The last term is due to the superposition of the two beams diffracted to either side of the lattice planes; it produces an additional period of $a/2$ and is independent of the coordinate z of defocussing under the limiting supposition (III) made above.

In-focus image

First, let us consider the intensity distribution at the exact focus by taking $z=0$. As the ratio A_2/A_1 does not, then, depend on the thickness, the intensity distribution across the fringes remains unaltered with varied thickness, the fringe structure being determined by the diffracting power s only. Thus, the structures of images at exact focus may give also information on the diffracting power s at least when only one set of lattice planes is involved. As long as $s \leqq 0.25$, there occur only intensity maxima which coincide with the lattice planes i.e. with the maxima of the inner potential. If s exceeds 0.25, in addition to these principal maxima, second weaker ones occur in the middle between the principal ones, increasing in strength with increasing s. For s approaching 1, naturally the assumptions made here fail because higher order diffractions occur at once together with the first order ones. (About this, some remarks will be made in another paper (this volume, p. 104). On the other hand, if the thickness of the crystal reaches the value $D=D_0$ most favourable for highest contrast in the fringes, the difference (maximum-minimum) of the intensities in the fringes may already be equal to the primary intensity at $s=0.065$, and may be as high as 3 times the primary for $s=0.5$.

Out-of-focus images

With defocussing, the coefficient A_1 oscillates in a manner already known from wave optics. (Equal intensity distributions would appear, repeated after distances of t, alternately shifted by half the period a.) Therefore, by defocussing to a suitable degree, we can usually raise the coefficient A_1 to give the highest possible contrast, unless β is already $\pi/2$, i.e. $D=D_0$, (see eqs. (4) and (6)). Fig. 1 shows the dependence on crystal thickness of A_1 for two planes z=const. For the full curve, the exit surface is chosen, and for the dashed curve that plane in which the value A_1 becomes highest for the in-

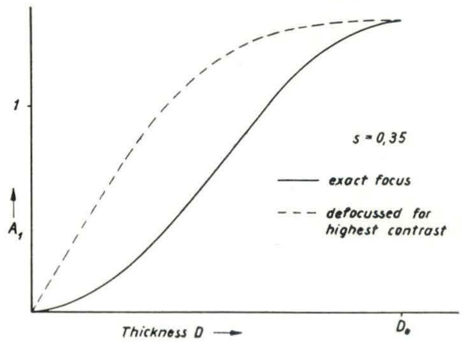

Fig. 1. Contrast coefficient A_1 from symmetrical diffractions of first orders.

dicated thickness. The nearer D is to D_0, the nearer is the plane of highest contrast to the exit surface, and the lower the available gain in contrast by defocussing to that plane. Whereas the curve for exact focus evidently reveals an "amplitude contrast" proportional to the square of thickness for very thin crystals, the difference between the two curves may be considered to represent an additional "phase contrast" which becomes visible at a suitable degree of defocussing. The latter, prevailing at small thicknesses, is gradually transformed into the former one as the radiation passes on through the crystal.

Conclusions

Some generalizations of the case assumed above, so as to include more general distributions of inner potential, and improvements by using Bethe's method to consider dynamical potentials, are easily possible if of the emerging beams only the first order diffracted ones have significant intensity. Then it can be deduced that several features in the behaviour of the intensity distribution vs. thickness and defocussing discussed above, are not peculiar to the assumptions made here, and would also exist if higher order diffractions are not neglected. Quantitatively, of course, these deductions will yet hardly allow any comparisons with the encouraging observations by Wyckoff and Labaw[1,2], Uyeda[3], Kamiya *et al.*[4] and Gansler and Nemetschek[5] where higher order diffracted beams surely take part in forming the microscope image. Such comparisons would require more generalized theoretical considerations, as well as more exact knowledge of the degree of defocussing and the thickness of the crystal in each observed case. It, therefore, seems to be premature to discuss some conformities and deviations of our deductions (peculiar of special assumptions) and the experimental observations referred to above.

At any rate, in contrast to the case in light microscopy, it will be of highest importance to achieve defocussed series connected with measurable defocus distances. Surely, it will not be easy (if at all possible in case of very small lattice spacings) to distinguish between images of the exit surface of an object and those of other planes nearby. The degree of defocussing may be determined from Fresnel fringes at the edges but with an uncertainty whose detrimental influence on the utilization of measured values increases with the resolution needed for small lattice spacings. After overcoming these difficulties, defocussed series would add information about the observed lattices, especially about their diffracting powers if several sets of lattice planes are involved. With suitable theoretical interpretation, they could provide an analysis of component waves, and perhaps, could compensate for missing small-area diffraction diagrams if the object consists of very small areas of single crystal structures.

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DISCUSSION

W. HOPPE: The condition of a sinusoidal electron potential, that means the absence of higher orders in the potential, will make it difficult to make experimental comparison.

H. NIEHRS: Surely you are right in that we have to consider also the higher Fourier coefficients of potential, if we want to compare theoretical deductions with experimental observations quantitatively. As long as we eliminate higher order diffracted beams, however, the inclusion of higher coefficients of the inner potential is of little importance. It does not alter the main features of the dependence on thickness and degree of defocussing of the image structure.

S. MIYAKE: About two years ago, I and Dr. Fujimoto realized the importance of taking into account many simultaneous reflexions when we consider the diffraction by a lattice plane with large spacing, and by using a tabulation of Mathieu functions we calculated numerically the intensities of different orders of reflexions, assuming a spacing in phthalocyanine crystal and value of the first order Fourier potential for it, and assuming a *sine-form potential* for the parallel incidence of electrons to the lattice plane. As the result, it was shown that the intensity of the first order reflexion is of course strong and that of the second order reflexion is also far from negligible.

H. NIEHRS: I agree with you in that we have to consider also the higher order diffracted beams when experimental observations in this field are to be explained. The results given here may be regarded only as a first step in this direction. The assumptions are simplified to give us an easy survey on what happens if the diffracting power, thickness or degree of defocussing are altered.