ELECTRON HOLOGRAPHY

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Electron holography was introduced in electron microscopy thirty years ago by Dennis Gabor (1948). Although the technique is only just beginning to prove itself useful it has been widely applied in other fields (B.J. Thompson, 1978). The attraction of holography is the possibility of reconstructing the complex amplitude of a wavefront from an intensity recording with diffraction limited resolution. Holograms also have the advantage of being taken first and imaged later, which minimises object exposure. Comprehensive discussions of electron holography are given by K.J. Hanssen (1973) and R.H. Wade (1979).

The method relies upon the interference of two coherent wavefronts, one of which is known. This reference wavefront is then used to illuminate the hologram which reconstructs by diffraction the unknown wavefront. A thorough treatment of the subject is given by R.J. Collier, C.B. Burckhardt and L.H. Liu (1971). With appropriate scaling the reconstruction can be made at any wavelength or by digital processing. A conjugate wavefront is also generated because the interference pattern records only the modulus of the phase difference. Let $U_R(x) = A_R(x)\, e^{i\phi_R(x)}$ be the complex amplitude of the reference wave, with a similar expression for the unknown image wave U_I, then the intensity at the hologram plane is given by

$$\begin{aligned} I_H(x) &= |U_R(x) + U_I(x)|^2 \\ &= A_R^2(x) + A_I^2(x) + 2A_R(x)\, A_I(x) \cos [\phi_R(x) - \phi_I(x)] \qquad (1) \end{aligned}$$

Interference fringes are made evident by the cosine term. In practice, there will be wavefronts within an angular spread and with a range of wavelengths. One may consider that wavefronts at each angle and each wavelength will form their own set of interference fringes, all overlapping (M.E. Haine and T. Mulvey, 1952, E.N. Leith and J. Upatnieks, 1967). The resolution of the hologram will be determined by the finest set of fringes that remain clearly defined when the different sets are added. This is quite comparable to the conclusions of the envelope approximation in contrast transfer theory (K.J. Hanssen and L. Trepte, 1971, J. Frank, 1973) where it is found, for small deviations from perfect coherence, that the effect is simply to multiply the transfer function by an envelope that attenuates higher spatial frequencies, thus eliminating finer fringes. The highest resolution is only attainable about a certain small defocus (R.H. Wade, 1978, J. Frank, 1978) in conventional transmission electron microscopy.

Before reconstruction the hologram is processed so that its transmittance, $t_H(x)$, is proportional to the intensity to which it was exposed, i.e. $t_H(x) = g\,I_H(x)$. Thus the reconstructed wavefront is given by $U_c(X) = U_R(x)\ t_H(x)$, or

$$U_c(x) = g|U_R(x)|^2\ U_R(x) + g|U_I(x)|^2\ U_R(x) + g|U_R(x)|^2\ U_I(x) + g\ U_R^2(x)\ U_I^*(x) \qquad (2)$$

The last two terms are generated by the interference between the reference and image waves. The third term is directly proportional to the original image wave and the fourth to its conjugate. However, $t_H(x)$ may have both an amplitude and phase component and the type of hologram used determines the quantity that is linearly reconstructed (K.J. Hanssen 1970, R.H. Wade 1974). If g is purely real or purely imaginary the complex amplitude reconstructed is proportional to that of the image wave, whose components may be separately visualised by optical methods such as interferometry or phase contrast (G.W. Ellis 1966, K.J. Hanssen 1973). For the special case of a weak phase object the intensity reconstructed from a weak phase hologram will be proportional to the original phase distribution, and similarly for a weak amplitude object.

The quantity $U_R(x)$ is determined by the layout. For a plane, tilted reference wave, $U_R(x) = e^{i\beta x/\lambda}$, where λ is the wavelength, and equation 2 becomes :

$$U_c(x) = g\ (1 + A_I^2(x))e^{i\beta x/\lambda} + g\ A_I(x)e^{i\phi_I(x)} + g\ A_I(x)e^{i(2\beta x/\lambda - \phi_I(x))} \qquad (3)$$

The inclination of the reference with respect to the image wave results in the spatial separation of the reconstruction and its conjugate from the illuminating beam.

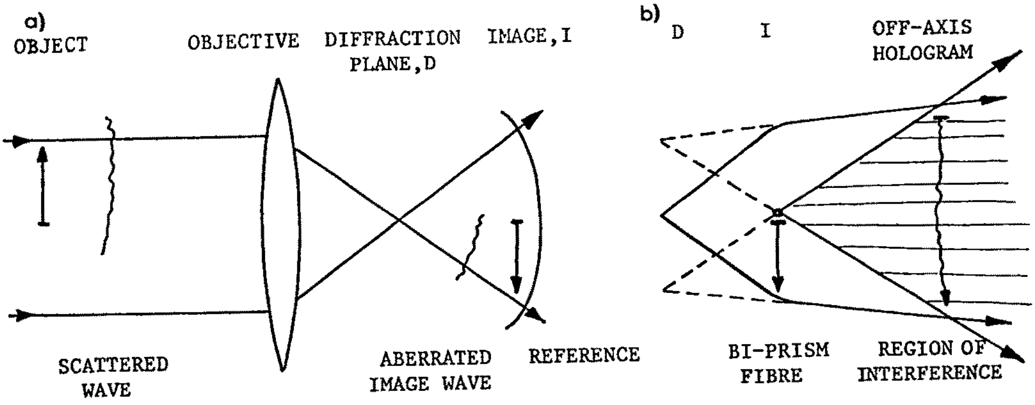

Figure 1 : Holographic layouts in electron microscopy. a) In-line. The unscattered illumination acts as a reference wave. b) Off-axis. The bi-prism produces interference between the image and an adjacent clear area.

This can be seen clearly from the spectrum of the hologram,

$$\tilde{I}_H(u) = \delta(u) + \tilde{U}_I(u) \otimes \tilde{U}_I(u) + \tilde{U}_I^*(u-\beta/\lambda) + \tilde{U}_I\,(u+\beta/\lambda) \qquad (4)$$

thus the reference beam inclination angle β should be large enough to separate the off-axis terms from the central auto-correlation. In general, this implies $\beta > 3\, u_o\lambda$, where u_o is the highest frequency in the image, but the special case of a weak object requires only that $\beta > 2\, u_o\lambda$ since the auto-correlation is limited to the bias.

If in recording the hologram the reference and image wave are on the same axis, i.e. $\beta = 0$, then the reconstructions will be in-line and all the terms of equation 4 will be centered on the origin. In-line holography is only appropriate for objects whose auto-correlation is negligeable and which transmit enough of the beam unscattered to act as a reference wave. Gabor (1949) originally proposed a projection method, using the change in wavelength of 10^5 between the electron microscope and the light reconstruction to provide magnification. The arrangement was too difficult to use at the time, although it can now be compared to S.T.E.M. imaging (L.H. Veneklasen, 1975) and was later shown by R.W. Meier (1965) to introduce additional aberrations. A typical brightfield micrograph however may also be interpreted as an in-line hologram, as shown in figure 1. To compensate for the original aberration and defocus the reconstruction is imaged backwards through a light optical model of the electron microscope (K.J. Hanssen, 1968, J. Rogers, 1978). At the original object plane is a perfect reconstruction and a conjugate having twice the image defocus and aberration, whose combined intensity may be described by an all positive transfer function which in general still contains zeros.

The first electron holograms were taken using the in-line method by M. Haine and T. Mulvey (1952). The specimens used were zinc oxide crystals and carbon black. Resolution at the time was about 10 Å and limited by mechanical and electrical instability. The resolution of the reconstructions did not match that of the holograms. Similar experiments were performed by T. Hibi (1956). The first optical reconstruction of an electron hologram that achieved the resolution of the original, 3 Å, was made by K.J. Hanssen and G. Ade (1976) using a carbon film as object with full com-

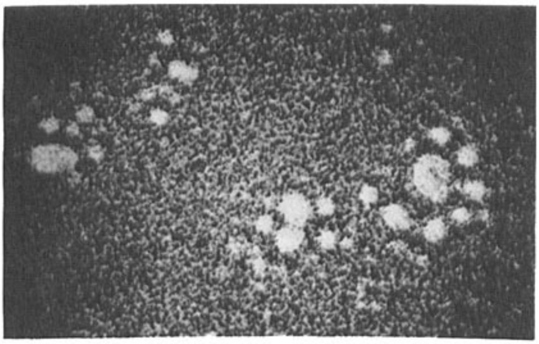

Figure 2 : Reconstructed image of gold particles on a carbon film from an in-line hologram recorded at a defocus of 2.1 µm and 10^5 magnification. The diffractogram extends to 7 Å. (from N. BONNET et al, 1978).

200 Å

pensation of spherical aberration and defocus. With contemporary microscopes the effects of spherical aberration are only noticeable at resolutions below about 5 Å.

When an in-line hologram is taken at a very large defocus, in the far-field of the whole image, the two reconstructions will be so far separated that one will be undisturbed by the other. The local resolution will be continuous, i.e. the transfer function will be constant and without zeros, although if the whole reconstruction is considered the effects of the superimposed conjugate image will still be apparent. Fraunhofer holography, as it is called, has been the subject of many experiments in electron microscopy e.g. A Tonomura et al. (1968), J. Munch (1975), M. Troyon (1977), N. Bonnet et al. (1978). Because of the need for high spatial coherence to record any detail at defocus values of the order of microns a field emission gun is essential. Even so the resolution will be limited by the source and only defocus need be compensated. The best reconstruction to date is that shown in figure 2 whose diffractogram extends to 7 Å.

Single-sideband holography uses a half-plane aperture in the diffraction plane of the microscope and has been thoroughly investigated despite great experimental difficulty, by K.H. Downing (1974). In general two complementary holograms are required, but the conjugate reconstruction is suppressed and in a fully compensated optical system a resolution of 4 Å was obtained. True off-axis holography is required to reconstruct a complex object from one hologram. The difficulty lies in introducing a coherent, tilted reference wave. The most practical method is to use the equivalent of the Fresnel bi-prism (T. Hibi and K. Yada, 1976) at the image plane, as shown in figure 1b. The first demonstrations of off-axis electron holography were by G. Mollenstedt and H. Wahl (1968) and by H. Tomita et al (1970) who

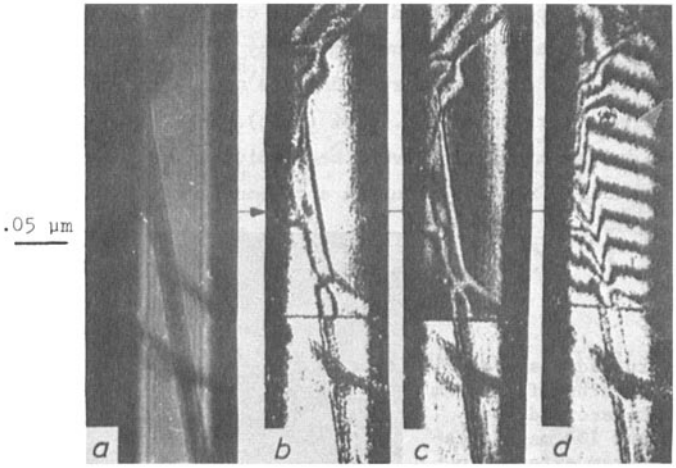

Figure 3 : Off-axis results from H. WAHL (1974). a) Hologram of ZnO crystals. b) -d) Reconstructions. The upper part is doubly illuminated for interference by a plane wave whose phase is b) 0 , c) π and d) slanted.

worked at a resolution of 500-1000 Å. An investigation of the method was made by G. Saxon (1972) with consideration of aberration correction. A more flexible microscope and bleaching the holograms before reconstruction enabled H. Tomita et al. (1972) to obtain an apparent resolution of 20 Å. Considerable development applied to improving the brightness and monochromaticity of the source has allowed A. Tonomura et al. (1979a) to obtain off-axis holograms in a microscope that has demonstrated .62 Å lattice fringes. A bi-prism working at up to 200 V produced a large field of 2 Å interference fringes at a defocus of 6000 Å. Since the objects were pentagonal gold crystals about 100 Å across the hologram is already in the Fraunhofer field. A through focus series of the optical reconstruction, which is not compensated for spherical aberration, clearly shows 2.4 Å lattice fringes and their half-spacing.

The three dimensional nature of the reconstruction was shown by J. Munch (1975). H. Wahl (1974, 1975) demonstrated with electrons interferometric techniques developed in light optics to produce the difference between two image waves or interference between reconstructions from two holograms, as shown in figure 3. The resolution was limited to about 50 Å and the specimens, as in most previous work were crystals of zinc or magnesium oxide. The technique has recently been applied to the study of magnetic domain walls (B. Lau and G. Pozzi, 1978). A Mach-Zehnder reconstruction of a hologram obtained by A. Tonomura et al. (1979b) clearly demonstrates the practicality of such methods for phase visualisation.

The continuous resolution obtained from most off-axis holograms has been much less than from in-line techniques. This is because a low magnification is used to balance the conflicting demands of a small source size and sufficient intensity for a short exposure. But such technical difficulties can be overcome, as shown by the work of A. Tonomura et al. Aberration compensation must be used in the reconstruction to obtain the best possible results. With an exposure time of a few seconds off-axis holography may now be applied to more delicate specimens. However there are not many microscopes that could meet this standard at present. Hence for most high resolution work in-line techniques offer the simplest approach to image interpretation.

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