Sensitivity-Enhanced Electron-Holographic Interferometry and Thickness-Measurement Applications at Atomic Scale

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An electron phase shift down to an order of $\lambda/100$ (λ is wavelength) was detected for the first time. This was realized by applying longitudinally reversed shearing interferometry in the optical reconstruction stage of electron holography. Thickness changes due to monatomic steps in MoS_2 films were clearly observed in phase-amplified interference micrographs.

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Interferometry renders the phase information of a wave observable. In the optical case, the minimum detectable phase shift reaches down to $\lambda/1000$ (λ is wavelength), and consequently even a thickness change of a few angstroms can be detected. In electron interferometry using a Möllenstedt-type biprism,[1] however, the minimum detectable thickness change has been 100 Å at best, in spite of the extremely short electron wavelength. This is because the refractive indices of the specimens are so close to that of a vacuum. If electron phase shifts as small as $\lambda/100$ become detectable, measurements on a truly atomic scale, not only in the thickness direction but in three dimensions, can be practical when combined with electron microscopy.

In recent experiments[2] conducted by the authors using electron holography,[3] interference micrographs phase amplified ten times were obtained, where contour lines were drawn in $\lambda/10$ units. This method employs a higher-order diffracted beam and its conjugate utilizing the film's nonlinearity.[4] The phase-shift value of $\lambda/10$ is the smallest previously detected, and corresponds to a thickness change of about 30–40 Å and a magnetic flux[5] of $h/10e$ in interference microscopy.

In the present experiment, a successful attempt to detect an electron phase shift of order $\lambda/100$ was made using another phase-amplification technique which does not rely on film nonlinearity. The experimental procedure consists of two steps: electron hologram formation and optical reconstruction. Electron holograms were formed with our recently developed field-emission electron microscope,[6] in which an electron biprism was installed. A schematic diagram of it is shown in Fig. 1. A specimen is situated only in one-half of the specimen plane. Its image is formed through an objective lens. The biprism makes the specimen image overlap with the reference wave passing beside the specimen. Special attention was paid to forming the holograms so that the peak positions of the recorded fringes can be recorded with an accuracy of 1/100 of the fringe spacing. That is, the spatial coherence length of the illuminating electron beam was adjusted so that it was nearly 100 times larger than the minimum coherence length to form carrier fringes. In addition, the fringe spacing was set to 100–200 μm,

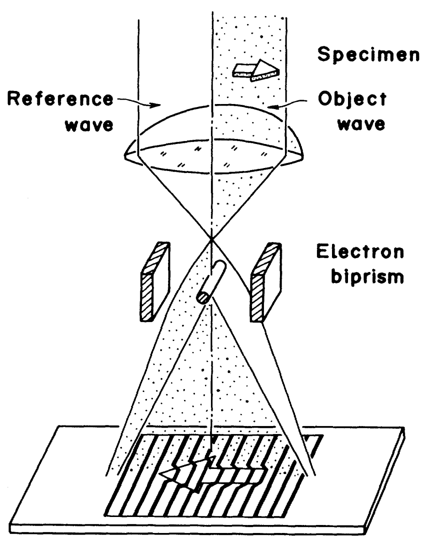

FIG. 1. Schematic diagram for electron hologram formation.

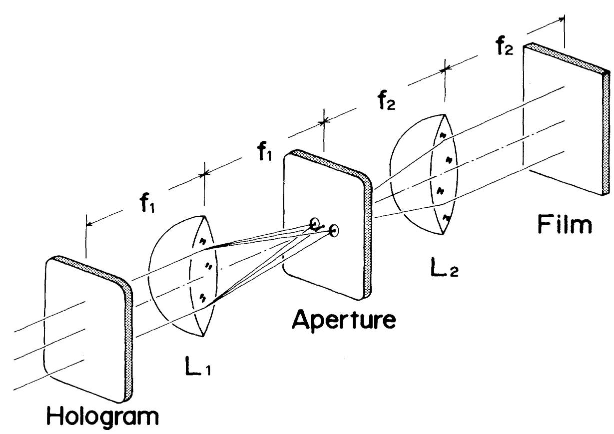

FIG. 2. Optical spatial filtering setup for forming phase-amplified holograms.

which is more than 100 times larger than the resolution of the recording medium, Kodak High-speed Holographic Plate 131. The specimen magnification was 5000 times, and the electron accelerating voltage was 45 kV.

Phase amplification of the electron hologram was carried out with optical spatial filtering as shown in Fig. 2. This technique was first proposed in the optical field by Bryngdahl[7] and developed by Matsuda, Freund, and Hariharan.[8] A collimated laser beam illuminates the hologram and only the first diffracted spots are selected. This is done with an aperture which has two holes situated at the back focal plane of lens L_1. Then, a hologram phase amplified two times is formed and recorded on a film. The film is bleached to get higher diffraction efficiency. When this procedure is repeated n times, the amplification rate is 2^n times.

Phase-amplified interference micrographs can be obtained from phase-amplified holograms using the optical system shown in Fig. 3. A collimated laser beam is split in two by a Mach-Zehnder-type interferometer. The two beams illuminate a hologram, producing two sets of a reconstructed image and its conjugate. Only the reconstructed image of one beam and the conjugate image of the other beam pass through the aperture. An interference micrograph phase amplified two times can be obtained. Thus, an overall amplification of 2^{n+1} times is obtained. The amplification rate may be made as high as desired with this procedure, but the problem is whether it is possible to actually read out a phase shift as small as $\lambda/100$ from the amplified interference micrographs.

To test the possibilities of detecting such small phase shifts, cleaved thin films of molybdenite (MoS_2) crystal were used as specimens. A well-prepared surface of this crystal has atomically flat surfaces and well-defined atomic steps, and therefore the specimens present phase objects with given phase variations of order $\lambda/100$. The thickness change producing a phase shift of one wavelength is given by $2\phi_0\lambda/V_0$, where ϕ_0 and V_0 are the accelerating voltage of the electron beam and the inner potential of the specimen, respectively. The thickness is calculated to be 300 Å when the inner potential is assumed to be 16.3 V. Therefore, a thickness change of 6.2 Å (one-half of the c-axis lattice spacing[9]) produces a phase shift of 0.02λ.

Specimens were prepared by cleaving molybdenite crystal as thin as possible with tweezers, and only films with a nearly uniform thickness of less than 50 Å were selected. Then, the films were placed on copper meshes. An electron micrograph of molybdenite film is shown in Fig. 4. Although extinction contours due to Bragg reflections are often observed as shown in the micrograph, only film regions where no Bragg reflection was excited were selected. This is because the phase of the transmitted electron beam depends on the Bragg conditions,[10] which constitutes an obstacle to the absolute measurement of the electron phase.

Interference micrographs of molybdenite film are shown in Fig. 5. The amplification of the micrographs (a)–(d) are 1, 4, 8, and 12 times, respectively. No fringe shift can be perceived in the conventional micrograph shown in Fig. 5(a). However, the amplification increase makes it possible to observe surface steps in the micrographs. Fringe steps can be perceived along the two oblique lines designated A and B in the micrographs.

An interference micrograph amplified 24 times is shown in Fig. 6. Here, a phase shift of 0.025λ can also be recognized along the line designated C, which corresponds to a monatomic step of 6.2 Å. This identification is guaranteed by the fact that all shift values measured were integral multiples of this minimum value of 0.025λ. For example, the measured values of phase shifts at steps A and B in Fig. 6 are 0.125λ and 0.075λ, corresponding to five- and three-layer steps, respectively.

Fluctuations in interference fringes become appreciable with larger amplification. They are mainly due to granular noise in the recording media, the partial spatial coherence of the illuminating electron beam in forming holograms, and Fresnel fringes due to the edges of the biprism wire overlapping the hologram. Among these, the influence of the Fresnel fringes (see Fig. 7) is appreciable in the peripheral regions of a

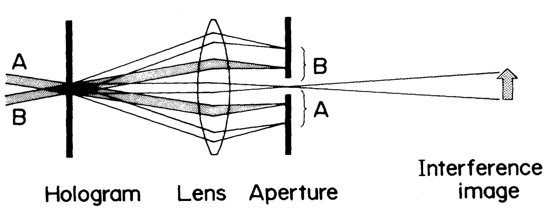

FIG. 3. Optical reconstruction system for interference microscopy.

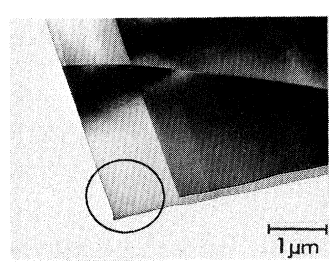

FIG. 4. Electron micrograph of molybdenite thin film. Only film regions where no extinction contours due to Bragg reflections appear are selected, as indicated by the circle in the figure.

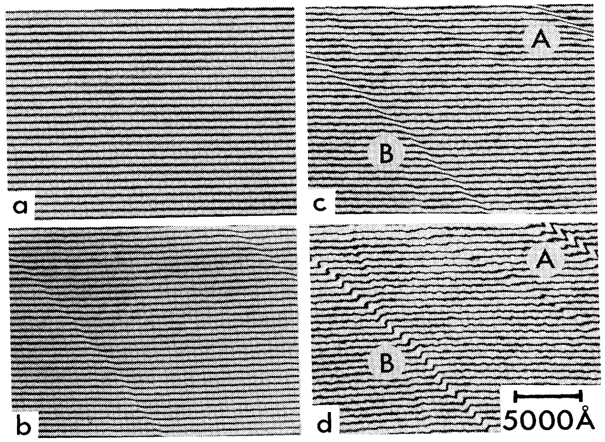

FIG. 5. Phase-amplified interference micrographs of molybdenite thin film. The amplifications in (a)–(d) are 1, 4, 8, and 12, respectively.

hologram. Therefore, phase measurements are restricted to the central part of a hologram, the only place where the Fresnel fringe intensity is weak enough for accurate measurements. The mean value of the total fluctuations in interference fringes is approximately $\lambda/100$, which limits measurement accuracy.

Conversely, this technique can be utilized for the precise measurement of the inner potential V_0, instead of the thickness distribution, of a specimen, since the height differences due to steps are definitely known. In our case of MoS_2, the measured value of V_0 was 20 ± 2 V. Although this value is a little different from the theoretical value of 16.3 V calculated from atomic scattering factors, it is fairly consistent with the measured values of 17.1 V[11] and 19.5 V.[12]

In this experiment, it was confirmed for the first time that an electron phase shift could be detected with a measurement accuracy on the order of $\lambda/100$ by means of electron holography. This technique can be applied to magnetic-flux measurements with an accuracy of $h/100e$ as well as thickness measurements of a few angstrom units. Although such thickness and magnetic-flux measurements are also possible through the use of light and superconducting Cooper pairs, respectively, electron holography is distinguished by image resolutions as high as a few angstrom units.

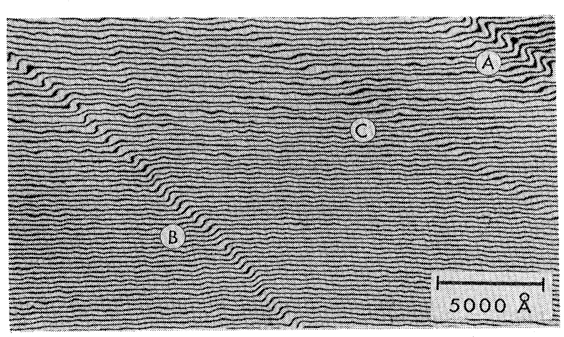

FIG. 6. Interference micrograph of molybdenite film phase amplified 24 times.

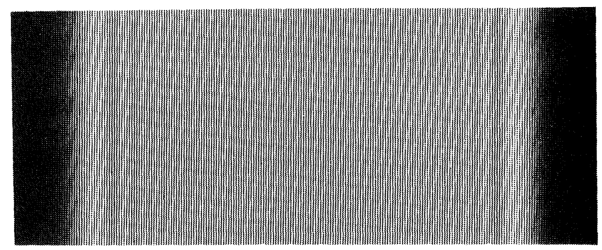

FIG. 7. Fresnel fringes overlapping the peripheral regions of a hologram.

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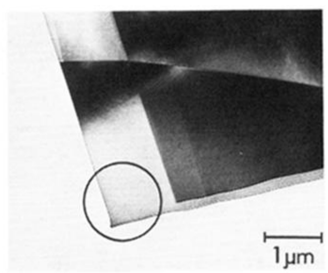

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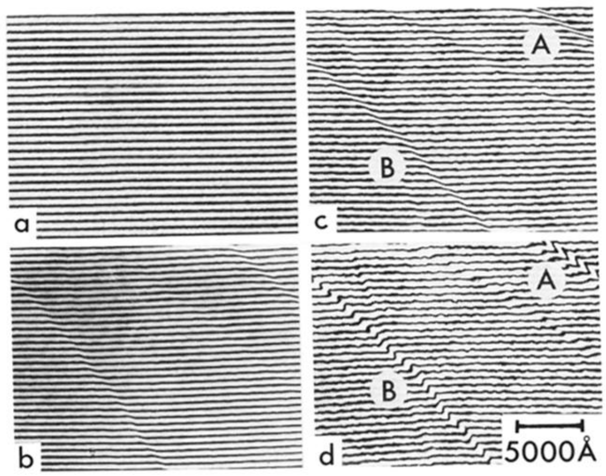

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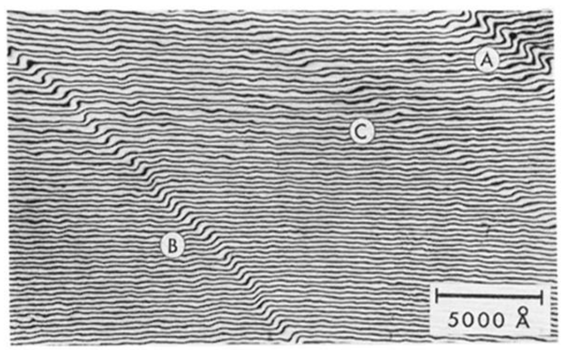

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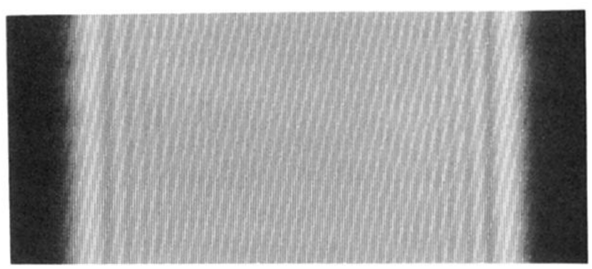

FIG. 7. Fresnel fringes overlapping the peripheral regions of a hologram.