OVERVIEW

Applications of Electron Holography to Materials Science

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The new measuring technique using electron holography was applied to various measurements of materials. Electron holography is a two-step imaging method. In the first step, interference image is formed in a field emission electron microscope by superimposing a reference electron wave on an object electron wave which transmitted through a specimen. In the second step, the optical image is reconstructed in an optical interferometer. Among several applications of this technique, interference microscopy in particular was proven to be promising for practical measurements, since this technique enables to measure phase distribution of an object wave by interference between the reconstructed image and a reference laser beam.

The distribution of fringes in an optical interference micrograph indicated the thickness distribution of a homogeneous specimen and the magnetic lines of force inside and outside of a ferromagnetic materials. This paper describes the experimental results such as distribution measurements of thickness and magnetization in thin films and fine particles, direct observation of quantized magnetic fluxes and surface morphology of crystal specimen.

The potentiality of this technique was clearly demonstrated and further developments of electron holography are expected to open new possibilities of observation and measurements in atomic dimensions.

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I. Introduction

Electron microscopy is used as an important part of measurement technology in such fields as physics, biology, medical and material sciences. The electron microscope was invented by Ruska⁽¹⁾ in 1933, to provide high resolution observation, as a substitute for light microscopes, in which resolution had already reached a theoretical limit. At that time, the electron microscope was believed to be able to resolve atom images, because the wavelength of an electron wave is two orders smaller than the size of an atom. The resolution quality has improved dramatically over the last 50 years, and in some cases, we can even observe atom images using one of the latest electron microscopes. But resolution is still not high enough in comparison to the electron wavelength. It is limited by aberrations of electron lenses.

In 1948, Gabor⁽²⁾ invented electron holography to achieve higher resolution, without using any electron lenses at all. The method consists of two imaging processes. The first process is a recording of an interference image called a hologram by interfering two electron waves. The second is image reconstruction from the hologram by light illumination. Originally, an image of a specimen could only be magnified by the difference be-

tween the electron and the light wavelengths. Gabor was successful in only a fundamental experiment which used light waves in both processes, because he could not utilize a coherent electron wave. After his experiment, some attempts concerned with creating electron interference were made⁽³⁾⁽⁴⁾, but still electron holography could not be put into practical use. The first success in creating interference was achieved by Tonomura *et al.*⁽⁵⁾ and Möllenstedt *et al.*⁽⁶⁾ in 1968. In their experiments, images of specimens were actually reconstructed, although their qualities were inferior to those of the original electron micrographs.

In 1978, an electron microscope in which a field emission electron gun was installed was developed by Tonomura *et al.*⁽⁷⁾ Using this instrument, more than 3000 interference fringes could be obtained. This proved that the coherence of electron waves could be improved by a factor of ten. Electron holography was applied not only to improvement in resolution⁽⁸⁾ but also to new measuring techniques such as interference and phase difference electron microscopes⁽⁹⁾. Among these techniques, interference electron microscopy was expected to be used as a new method in the material research field⁽¹⁰⁾. Since then, this technique has been applied many times. This paper describes the experimental results and potentiality concerned with interference electron microscopy employing electron holography.

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II. Electron Holography

As mentioned above, electron holography consists of two imaging processes. The first is hologram formation in an electron microscope, and the second is image reconstruction on an optical bench. These processes are shown in Figs. 1 and 2.

When a wave passes through or is reflected from a specimen, the amplitude and phase of the wave is modulated by the specimen. In a conventional electron microscope, an image is magnified and recorded on a photographic film. In electron holography, another electron wave, which is called a reference wave, is overlapped on the original image (the object wave) to form an interference pattern. This pattern is magnified by electron lenses and recorded on a photographic film as a hologram. The information on the specimen is recorded as local modulations in directions and spacings of the interference fringes. When a laser beam illuminates a hologram, it is deflected according to the direction and spacing of the fringes. In diffracted waves, two images, a real and a conjugate image are formed.

Figure 3 shows a schematic diagram of hologram formation in an electron microscope. Electrons emitted from a field emission source illuminate a specimen which occupies only half of the specimen plane. An electron biprism⁽¹¹⁾ is installed between the objective and the intermediate lens of the electron microscope. This device consists of a fine wire and two grounded electrodes as shown in Fig. 3. When a potential of 50–200 V is applied to the wire, two electron waves which pass through the

Fig. 1 Formation of electron hologram.

Fig. 2 Image reconstruction.

Schematic of interference electron microscope

Fig. 3 Hologram formation in electron microscope.

Fig. 4 Field emission electron microscope.

Fig. 5 Micrographs of a field emission source.

specimen and the empty space are overlapped and form an interference image.

Figure 4 shows a field emission electron microscope. The lower part of the column is almost the same as that of a conventional electron microscope, except for an electron biprism which is installed between the objective and the first intermediate lens. The upper part of the column has special features. The material of the outer shells of the illumination system is made of Permalloy to shield against alternative magnetic fields emanating from peripheral apparatus. The electron gun has a field emission electron source which is made of a fine tungsten $\langle 310 \rangle$ single crystal needle shown in Fig. 5. In this type of electron gun, electrons are emitted from a virtual source of about 5 nm without heat. In this microscope, electron coherence is improved because of the small source size and the monochromaticity of the beam.

III. Interference Electron Microscope

1. Principle of interference electron microscope

The phase distribution of an object wave is detected using interference microscopy. Figure 6 is a schematic diagram of an interference microscope. An electron hologram is set into a Mach-Zehnder interferometer which contains two half-mirrors and two mirrors. When a collimated laser beam illuminates the hologram, two images are reconstructed. One of them is formed on an observation plane by two lenses. An aperture is inserted on the back-focal plane of the first lens to remove the transmitted laser beam and the conjugate image.

The laser beam, which is split by the first half-mirror in the interferometer, is overlapped on the image to reveal

the phase distribution of the object wave. If the directions of the two laser beams coincide with each other, a contour map of the phase distribution can be observed.

2. Phase difference amplification

In an ordinary interference microscope, fringes in a contour map appear when the phase shift equals one wavelength. To overcome this limitation, two amplification methods based on a characteristic feature of holography were employed⁽¹²⁾.

The first method is shown in Fig. 7. A collimated laser beam is split by a Mach-Zehnder interferometer. Each beam illuminates an in-focus hologram from a slightly different direction. The first-order diffracted wave of beam B and the conjugate image of beam A are arranged so that two images are formed at the same position on an observation plane. The amount of the phase shift in these two images is the same but their orientations are reversed. Subsequently, the phase difference is amplified two

Fig. 6 Optical reconstruction system for interference microscopy.

Fig. 7 Optical arrangement for phase difference amplification (1).

Fig. 8 Optical arrangement for phase difference amplification (2).

times to provide the interference image. If an N th-order and a minus N th-order diffracted wave originating from a non-linear hologram interfere with each other, a $2N$ -times amplified phase distribution will be obtained⁽¹³⁾.

The second amplification method is shown in Fig. 8. This optical system has two imaging lenses which are arranged as a confocal system. A plate with two pinholes is set on the back-focal plane of the first imaging lens so

that the plus and minus first-order diffracted waves pass through them. These two waves form a two-times amplified hologram on a photographic film set on the observation plane. This phase-amplified hologram is photographically processed and is placed back as the object plane of the confocal system, in place of the original hologram. Then, a four-times amplified hologram is formed on the observation plane. If this process is iterated N times, a 2^N -times amplified hologram can be obtained. Finally, the 2^N -times amplified hologram is set in the optical system shown in Fig. 7, and a 2^{N+1} -times amplified phase distribution can be observed⁽¹⁴⁾.

IV. Thickness Measurement

When an electron beam is transmitted through a specimen, they are accelerated by the inner potential of the specimen. Consequently, the wavelength of the electrons becomes shorter and the phase of electron wave is delayed. The phase shift is expressed in the following formula:

Fig. 9 Interference micrographs of magnesium oxide particles. (a) Reconstructed image and (b) Contour map.

Fig. 10 Phase-amplified interference micrographs of magnesium oxide particles. (a) Interferogram and (b) Contour map.

$$\Delta\phi = \frac{\pi V_0 t}{E\lambda}$$

where V_0 and t are the mean inner potential and thickness of the specimen, E is the accelerating voltage, and λ is the wavelength of the electron beam.

Figure 9 is an image (a) and a contour map (b) of magnesium oxide particles. The acceleration voltage is 80 kV. The magnification is 40000 times. The thickness distribution cannot be measured from the reconstructed image, but the contour map describes the thickness distribution because contour fringes appear in every 50 nm thickness change. Detailed thickness distribution can

be observed in Fig. 10 in which the phase distribution was amplified two times. In this figure, continuous thickness changes (a) and fine contour lines (b) can be measured.

Figure 11 shows a reconstructed (a) and an interference (b) image of a beryllium particle⁽¹⁵⁾ on a carbon film. The conditions of hologram formation are the same as employed in Figs. 9 and 10. In this case, the thickness contours appear in every 46 nm thickness change. The two-times amplified contour map and schematic view of the particle are shown in Fig. 12. Figure 13 is a 32-times phase amplified contour map of the same specimen. This phase distribution was amplified by the iteration method

Fig. 11 Interference micrographs of a beryllium particle. (a) Reconstructed image and (b) Contour map.

Fig. 12 Phase-amplified contour map of a beryllium particle. (a) Two-times amplified contour map and (b) Schematic view of the particle.

Fig. 13 Phase-amplified contour map of a beryllium particle.

Fig. 14 Thickness distribution of a molybdenite thin film.

illustrated in Fig. 8. The contour fringes show 1.4 nm thickness changes.

Figure 14 shows thickness distribution of a molybdenite (MoS_2) thin film. The acceleration voltage is 45 kV. Phase amplification was performed by both the iteration and high-order interference methods. The amount of the phase shifts at A, B and C in the figure are 0.025, 0.075 and 0.125 wavelengths respectively. It is known that this material is cleaved at intervals of 0.62 nm⁽¹⁶⁾. The thickness change at A in Fig. 14 is calculated as 0.66 nm; this value is in close agreement with that determined by crystallography. If the height of the step is found to be 0.62 nm, the mean inner potential value is calculated as 21 V.

V. Magnetic Field Measurement

We assume that two electron waves which diverge from a single point pass through a magnetic field and converge to a single point. The phase difference between two waves caused by the magnetic field is expressed by the following formula:

$$\Delta\phi = \frac{2\pi e}{h} \int_S \mathbf{B} \cdot d\mathbf{n}$$

where h is Planck's constant, $\mathbf{B}$ is magnetic field strength, S is the surface whose perimeter is defined by the trajectories of the two electrons, and $\mathbf{n}$ is a surface element vector which is perpendicular to the surface S . If the magnetic field $\mathbf{B}$ satisfies the following formula:

$$\int_S \mathbf{B} \cdot d\mathbf{n} = \frac{h}{e} = 4.1 \times 10^{-15} \text{ [Wb]}$$

the phase difference becomes equal to one wavelength. This fact means that a constant magnetic flux of h/e flows between two adjacent contour lines. The directions

Fig. 15 Observation of recorded magnetization pattern.

of the magnetic lines of force can be determined from those of the interference fringes⁽¹⁷⁾.

Figure 15 is an example of a magnetic field observation. The magnetic field was recorded by a moving magnetic head on an evaporated cobalt film. The interference contour fringes along the arrows indicate directions of recorded magnetization inside the film. In the lower part of the photograph, leakage flux from the film can also be observed.

Figure 16 shows interference micrographs of an plate-shaped particle of fcc cobalt. In Fig. 16(a), no contrast can be observed inside the particle. However, in (c), concentric contour fringes appear and we can observe the magnetic lines of force inside the specimen. The rotation direction of the magnetic lines of force is revealed by the interferogram (b). The lateral fringes shift downward at the edge of the specimen, then go upward into it. The direction of an electron phase shift caused by magnetization is reversed in orientation compared to that caused by specimen thickness. Therefore, the direction of magnetic lines of force is determined to be clockwise.

An example of phase amplification is shown in Fig. 17. The specimen here is also a cobalt particle. In this case, only one magnetic line of force appears in the ordinary contour map (b), because the sample is very thin. The phase-amplified contour maps (c, d, e) were made using

the interference between high-order diffracted waves. Detailed magnetization distribution is revealed in photographs (c) and (d). But in (e), image quality becomes quite poor, and some portions of the magnetic lines of force are no longer observable. This means that the coherence of third- or fourth-order diffracted waves is not high enough to form interference fringes in this experiment.

Interference microscopy was also applied to a superconducting material⁽¹⁸⁾. Figure 18 shows interference micrographs of magnetic fluxes which penetrate superconducting lead films. The lead films were fabricated on one side of a tungsten wire about 30 μm thick by a vacuum evaporation method. Prior to the evaporation, the tungsten wire was flash-heated by electric current to provide a clean and smooth surface. The critical temperature of the film was 7.2 K. This specimen was set into a low temperature specimen stage installed in a field emission electron microscope. An electron-optical arrangement for hologram formation is shown in Fig. 19. A lateral magnetic field of about 10^{-4} Tesla was applied perpendicular to the film, and then the film was cooled to 4.5 K. In Fig. 18(a) and (b), the thickness of the films was 0.2 μm and 1 μm respectively. When the film thickness was 0.2 μm , the strength of the magnetic field which penetrated the film was found to be $h/2e$. This value cor-

Fig. 16 Interference micrographs of a cobalt particle. (a) Reconstructed image, (b) Interferogram, and (c) Contour map.

Fig. 17 Interference micrographs of a cobalt particle. (a) Reconstructed image, (b) Contour map, (c) Phase-amplified contour map ($3\times$), (d) Phase-amplified contour map ($5\times$), and (e) Phase-amplified contour map ($7\times$).

Fig. 18 Interference micrographs of magnetic fluxes penetrating superconducting films. Film thickness: (a) $0.2\ \mu\text{m}$ and (b) $1.0\ \mu\text{m}$.

Fig. 19 Electron-optical arrangement for hologram formation.

responds to a theoretical one for single fluxon. In Fig. 18 (b), the magnetic field penetrated in the form of bundles of several fluxons.

VI. Measurement of Surface Morphology

An electron holography technique suitable for surface investigation has recently been developed⁽¹⁹⁾. In this

Fig. 20 Electron-optical arrangement for reflection holography.

method, a reflected electron wave originating from the specimen surface is used to form a hologram. Figure 20 illustrates the electron-optical arrangement for this technique.

In electron holography using transmitted electron waves, we can only observe the thickness distribution in a specimen. On the other hand, the new reflection method reveals the surface undulations of a specimen. Sensitivity to thickness changes at the surface is two orders higher than that obtained by the transmission method.

Figure 21 presents an example of reflection electron

Fig. 21 Surface undulation of a GaAs single crystal. (a) Interferogram and (b) Calculated surface topography.

holography. The specimen is a single crystal of GaAs (110). The surface undulations around the screw dislocation can be directly measured by the shifts of the tilted interference fringes. A fringe shift which corresponds to one fringe spacing means a vertical displacement of 0.05 nm.

VII. Conclusions

The potentiality of interference electron microscopy was demonstrated. The characteristic features of this technique are summarized as follows:

- (1) Easy interpretation of thickness and magnetization distribution
- (2) Quantitative measurement with high sensitivity
- (3) Unrestricted amplification of phase shifts.
- (4) Simultaneous observation of an image and contour fringes.

These features give rise to expectations that interference electron microscopy will become a powerful tool for material investigation.

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