Coherent electron interference from amorphous TEM specimens

Rodney A. Herring1,*, Koh Saitoh2, Nobuo Tanaka2 and Takayoshi Tanji2

1CAMTEC, MENG, University of Victoria, British Columbia, Canada V8N 4T6 and 2EcoTopia, Nagoya University, Nagoya, 461-8603 Japan

*To whom correspondence should be addressed. E-mail: rherring@uvic.ca

Abstract

For the first time, the electron intensity on the diffraction plane from amorphous transmission electron microscope (TEM) specimens has been found to have sufficient coherence to produce fringes in interferograms that were created using a wavefront splitting method of diffracted beam interferometry. The fringes were found to exist from low to high electron-scattering angles. Their spatial frequency depended on the angular overlap of the interfering beams, which was controlled by an electron biprism. From these interferograms, phase information of amorphous materials, which is information now lacking and required for determining their atomic structures, was obtained. An immediate application of this interference is a new method to determine the spatial resolution of the TEM that occurs at the shear angle for fringe disappearance.

Keywords

amorphous materials, electron interferometry, phase information

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Introduction

Amorphous materials (a-materials) make up a large fraction of the known materials with most being scientifically and technologically important. There are many types with broad classifications including a-metals, a-semiconductors, some ceramics, glassy materials, most plastics and biological materials such as cells. Their atomic structures are complex, and for the most part progress in solving their structures has been slow [1]. This is primarily due to their structure factors lacking the required phase information. High-resolution images of amorphous materials reveal short-range order but no other details of their structures [2–4]. Diffraction reveals speckle patterns and diffuse rings against a highly diffuse background giving angle and intensity information but not their phase. Perhaps, one of the most developed methods to study amorphous materials is fluctuation electron microscopy [5–8], which is a dark-field, hollow-cone method taken as a function of probe position across the specimen that provides a statistical measurement of medium range order at the nanometer scale of the amorphous material.

Methods of electron holography [9] and electron interferometry have been used to measure the phase of materials, which is possible at the atomic scale, with holography giving the absolute phase and interferometry giving the relative phase. Off-axis electron holography [10], which produces holograms with the specimen in focus, is the most popular method employed [11,12]. For a-carbon films, it has revealed the film’s thickness, carbon’s mean inner potential and possibly the presence of an electrostatic surface charge [13] but not the phase related to the a-carbon structure.

Recently, two methods of interferometry/holography that interfere electron beams on the diffraction plane have been developed to measure the phase of crystals using convergent beams and planar beams [14,15] but not applied to amorphous materials as the carrier spatial frequency of the fringes in these interferograms were given by the angle defined...
by the Bragg diffracted beams [16]. A useful feature of producing interference on the diffraction plane is the separation of the electron intensity into its components represented by diffuse scattering and Bragg diffraction as a function of scattering angle. On the image plane, all of this intensity is overlaid, making it difficult to interpret. The interference of the electron intensity on the diffraction plane by diffracted beam interferometry (DBI) has found that most of the electron intensity representative of elastically and inelastically scattered electrons has sufficient coherence to form fringes when self-interfered [17,18]. Thus, the electron intensity from amorphous materials should also have sufficient coherence to form fringes when self-interfered. Previous attempts to create DBI interferograms from amorphous materials during the Tonomura Electron Wavefront project [19] failed likely due to the poorer stability of the early field emission gun electron microscopes and the lack of Cs correctors and imaging energy filters.

This article shows excellent interferograms containing the diffuse, speckle and ring intensity on the diffraction plane generated from a-carbon using a modern scanning/transmission electron microscope (S/TEM) equipped with Cs correctors and an imaging energy filter.

**Method**

DBI of amorphous materials was performed using a double Cs corrected JEM 2100F equipped with an electron biprism in the selected area aperture position and an imaging energy filter (GIF Tridiem). The JEM 2010F was operated at a voltage of 200 kV. The diffraction intensity was generated by the primary electron beam passing through the amorphous TEM specimen. An electron biprism placed below the specimen was used to interfere the diffracted intensity with its surrounding intensity as illustrated in Figs. 1 and 2. The biprism was rotatable, which enabled interference of the electron intensity over the entire 360° on the diffraction plane. The virtual sources of the amorphous intensities, S1 and S2, were created by a wavefront splitting the main beam, i.e. the 000g, by means of the electron biprism. Required for seeing fringes is a sufficiently long camera length, ~1 m, and for the electron biprism to be brought into focus on the diffraction plane, which was made possible by either changing the objective lens current or mechanically by changing the specimen’s eucentric height, which change the source plane from Z0 to Z1 with the biprism located on plane ZOB in Fig. 1b. For these experiments, the objective lens current was usually increased to produce an over-focus (Z0–Z1) of ~2 µm. Condenser aperture sizes of 30 and 40 µm were used to give shallow convergence angles, αc, of ~1 mrad. The biprism below the specimen intercepted the electron intensity as is seen on the diffraction plane creating a discernible shadow of itself. Thus, a minimum biprism voltage is required to overlap the electron intensity over the biprism’s shadow, which was ~12 V for these experiments. The electron intensity extending from the optic axis to high scattering angles, β, (~6 mrad), were able to be interfered. This large angular range enabled the diffuse, speckle and amorphous intensities to be interfered (Fig. 2). The spatial frequency of the fringes in these interferograms depended on the shear angle, αB, of the overlapped intensity, which depended on the voltage applied to the electron biprism that provided substantial control of the interference width and the fringe’s spatial frequency, making this method of interference possible over a wide range of beam convergence angles.

The interferograms were recorded by a GIF CCD camera requiring short collection times of ~0.1–0.5 s for the low-angle scattered electrons due to their high intensity coming primarily from the main beam and having high-contrast fringes. Longer collection times of ~0.5–2 s were required for the high-angle scattered electrons where the highest intensities came from the amorphous rings. The highest contrast fringes were found when using the zero-loss electrons of the GIF having an energy window of 5 eV. DigitalMicrograph was used to collect, record and reconstruct the interferograms with the latter using its Holoworks subroutine.

In these interferograms, the fringe spacing decreased as the biprism potential increased. For the geometry provided in Fig. 1b, the fringe spacing, Y, is given by

\[ Y \propto \frac{(a + b)}{2a\alpha_B} \]  

(1)
and its interference width, $W$, is given by

$$W = 2\left|\frac{a + b}{a}\right|\left(\alpha_B - \frac{ab}{a + b} - R\right)$$

where $\alpha_B$ is the deflection angle of the beam due to the voltage applied to the biprism, and $R$ is the biprism’s radius [20]. The camera length of the microscope was made large to have enough pixels per
fringe for their recording. To observe fringes over larger scattering angles, the camera length was decreased accordingly.

For the conditions where the amplitudes of the interfering beams on either side of the biprism are equal, $A_R = A_L$; the contrast or visibility of the fringes, $V$, is given by the maximum intensity, $I_{\text{max}}$, and minimum intensity, $I_{\text{min}}$, of the fringes,

$$V = \frac{I_{\text{max}} - I_{\text{min}}}{I_{\text{max}} + I_{\text{min}}} \quad (3)$$

For the condition where the contrast of the fringes were not equal, i.e. $A_R \neq A_L$, which is the more likely scenario for the speckle interference as depicted in Fig. 2, their degree of coherence, $\gamma$, is given by,

$$V = \frac{2|A_L||A_R|}{|A_L|^2 + |A_R|^2} |\gamma| \quad (4)$$

As is well known from light optics [21], the visibility of interference fringes depends on the source size, and $V$ is a measure of the spatial coherence of the source and, therefore, of the source size. If $A_R$ and $A_L$ are the intensities emitted by sources, $S_1$ and $S_2$ (Fig. 1b), they will separately produce a point on the observation plane. Where the waves interfere, the resulting intensity will be (ignoring diffraction effects),

$$I = A_1 + A_2 + 2\sqrt{AA_2}|\gamma|\cos\chi \quad (5)$$

where $\chi$ is a linear function of the path length only. For a slit source of width, $\delta$, which is appropriate for the biprism used for this experiment, $\gamma$ is given by [22],

$$\gamma = \frac{\sin(\pi\delta/\sigma)}{\pi\delta/\sigma} \quad (6)$$

where

$$\sigma = |Ya/b| = \lambda(a + b)/2\alpha_bb. \quad (7)$$

The measurement of spatial coherence by electron interferometry is expected to have an accuracy of about 4–5% and believed to be slightly better than that obtained by the analysis of Young’s fringes [20] produced by superimposing two micrographs of an amorphous specimen in an optical diffractometer [23]. Since high frequency fringes are produced by this method, a further enhancement is to Fourier transform the interferograms to determine the disappearance of the fringes. This will help apply this method of interference for measuring the spatial coherence of the microscope.

**Results**

DBI interferograms were produced from several types of amorphous materials including a-Ge, a-Si, a-C, a-Ti, a-GaAs, a-metal (Zr$_x$Ni$_y$Al$_z$) and SiO$_2$. For this paper, results from the a-C specimen are pre-
sented since a-C was the first material found to have intensities with sufficient coherence to form fringes. All interferograms used 200 keV zero-loss energy-filtered electrons having a 5-eV window. The camera lengths in Figs. 3, 4 and 5 are estimates since free lens imaging of the diffraction plane was used and reference angles using the amorphous diffraction intensity is too broad for an accurate measurement. Figure 3 shows a-C’s typical amorphous speckle and amorphous contrast out to the first amorphous ring that is being partially blocked by the shadow image of the biprism on the diffraction plane with no voltage applied. Since the biprism splits the beam, diffraction of electrons off its edges occurs perpendicular to the biprism’s length. Figure 4 shows an example of an interferogram having high-contrast fringes close to the 000 beams that were wavefront split, producing two high-intensity disks seen as the two bright disks. Reconstructing this interferogram into its phase and amplitude is shown in Fig. 4c and d, which showed good phase information resulting from the fringe shifts that are seen left of the symbol A, in and around the amorphous intensity. The carrier spatial frequency of the fringes depended on the applied voltage of the electron biprism, which was used to vary the overlap angle of the electron intensity. As seen in Fig. 5 for a biprism voltage of 70 V, the higher angles of beam overlap produced fine fringes with low contrast. The maximum angular overlap to produce fringes, which was verified by producing Fourier transforms of the interferograms, was ~120 V, which corresponds to an angle of ~8 mrad. At the plane of the virtual source, it corresponds to ~0.1 nm. Fringes in these DBI interferograms were found extending from low angles out to the amorphous ring of a-C and even beyond (Fig. 6). Extending out to high scattering angles, the highest contrast fringes were found in the areas of highest intensity. Currently, it is not clear whether the intensities that are producing the fringes in these interferograms are solely self-interference, e.g. speckle intensity with speckle intensity, ring intensity with ring intensity or mixed. It is possible that some coherent interference exists between the speckle intensity and the diffuse background intensity where they have lost the same amount of energy to the specimen. In fact, there are many possible combinations of elastically and inelastically scattered electron intensities interfering. To determine conclusively which intensities are responsible for the formation of the fringes would require an in-depth study. Figure 7 shows a good enhancement of the fringe contrast when the imaging energy filter using the zero-loss electrons was used.

The formation of fringes in the speckle and ring intensities was found for a large range of illumination area on the specimen. The size of the beam illuminated on the specimen depended on the focusing of the electron beam. For a focused beam, a spot size of ~0.3–0.4 nm was produced, which spread out the amorphous intensities over a large area making them highly diffuse as seen in Fig. 6, whereas, for a defocused beam, a spot size of hundreds of nanometers was produced, which made the amorphous intensities smaller and more concentrated as seen in Fig. 7.

The thickness of the specimen was not measured for these experiments. During the experiment, the best condition for producing the speckle and ring intensities while in diffraction was sought. Typically, for diffracted beam interference, this corresponds to a specimen thickness of one extinction distance or less, which, if we use the first amorphous ring corresponding to the 111 diffraction vector of diamond, can be approximated by ≤50 nm.
Discussion

For the first time, the electron intensities generated by amorphous materials has been shown to have sufficient coherence to form fringes, which was made possible using the DBI method of electron interference on the diffraction plane. It should be noted though that the phase information collected by these experiments is very complicated. We are not even sure whether self-interference, i.e. speckle intensity with speckle intensity, is solely responsible for the coherent interference as there are many sources of intensities interfering from the elastically and inelastically scattered electrons. To obtain the pure phase of the structure of amorphous materi-

Fig. 4. An interferogram of a-C using 200 keV zero-loss energy-filtered electrons having a 5-eV window and a 30-V biprism voltage. The interferogram in (a) has been contrast enhanced to more easily see the fringes, although the intensity of the split zero beam is very high. The interferogram in (b) comes from (a) with an increase in magnification of the area between the dash lines. And, (c) and (d) are the phase and amplitude reconstructions of (b), respectively, showing phase information to the left of the letter A obtained from the central area resulting from the shifts of the fringes seen in (b).
als, some sophisticated analysis is required. The benefit of using an imaging energy filter to get high-contrast fringes was also clearly shown.

For the DBI method of interferometry on the diffraction plane, this is the first time that the specimen was not used as the beam splitter, i.e. an amplitude beam splitter, to generate the virtual sources of the interfered beams. Instead, the biprism is used as a wavefront beam splitter, which has its advantages and disadvantages. An advantage is that the interference angle can be continuously varied using the biprism’s voltage, which enables many spatial frequencies to be produce with the amorphous intensities. A disadvantage is that a minimum voltage is required to begin the interference of the two beams, which for a thick biprism or poorly placed electron biprism in the microscope’s electron optics may prevent the observation of the fringes as the fringes may be too fine to be seen. If the fringes cannot be seen, one can try changing the observation plane using the intermediate lens current or reducing the biprism’s defocus using the eucentric height or objective lens current, which produces thicker fringes.

The fact that the fringes continue to form while the interference angle increases due to lateral coherence may enable measurement of the properties of quasi-particles such as phonons, plasmons, etc. in amorphous materials, which is one of the goals of DBI [17]. For such measurements, the effects of dynamic inelastic electron diffraction must be considered [24]. The biprism’s shadow on the diffraction plane requiring a minimum voltage for interference may limit the detection of low-frequency information, similar to off-axis electron holography on the specimen image plane [25].

In the short term, application of DBI of amorphous materials to measure the spatial resolution of an electron microscope appears to be possible. The maximum shear angle of the interfering beams on the diffraction plane corresponds to the maximum spatial resolution on the image plane by a Fourier transform. The shear angle for fringe is easily reached by changing the voltage of the electron biprism, which has a simple linear transfer function for beam deflection. If given a sufficiently thin biprism (~0.1 µm), a measurement can be made at any desired image defocus condition of the specimen. A challenge for developing this method is finding a suitable amorphous material that is stable. Structural changes due to electron beam damage and beam heating as well as surface charges can be problems, which are associated with a-C, a-Si and a-Ge. A candidate material is a-metal \((\text{Zr}_x \text{Ni}_y \text{Al}_z)\), which to date has been found to be the most stable, although crystallization effects due to structural instabilities may limit these materials as well.
Although much work is still required, this contribution to the characterization of amorphous materials is considered a good beginning to obtain phase information associated with amorphous materials that may ultimately lead to the determination of the now hidden details of their complex structures.

**Conclusions**

A new method of DBI interference involving amorphous materials, for the first time, has shown that the amorphous intensity has sufficient coherence to form fringes in its interferograms. These fringes extended from low to high electron-scattering angles and have spatial frequencies dependent on the overlap angle of the two interfering beams that is determined from the applied voltage on the electron biprism. The fringes discontinue to form as the shear angle of the interfering beams became large, which enables a new method to measure the spatial resolution of the microscope.

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