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Energy-Filtered Electron Holography

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When electrons pass through a material, they can pass through without losing energy such as elastically scattered electrons or they can lose or gain energy by inelastically scattering with the material's electrons. The elastically scattered electrons have been used in the simulations of lattice images, which are used to help determine the atomic structure of materials. Inelastically scattered electrons were ignored in the simulations because it was believed that they did not have the required coherence to interfere with themselves and they contributed only to the background intensity. Recently though, a great deal of interest has been generated in knowing whether the inelastically scattered electrons can also contribute to the lattice images in order to help explain the Stobbs factor [1], where the contrast in the lattice images is often three or more times less than the theory predicts. The Stobbs factor makes it difficult, if not impossible, to establish quantitative values to high-resolution lattice images.

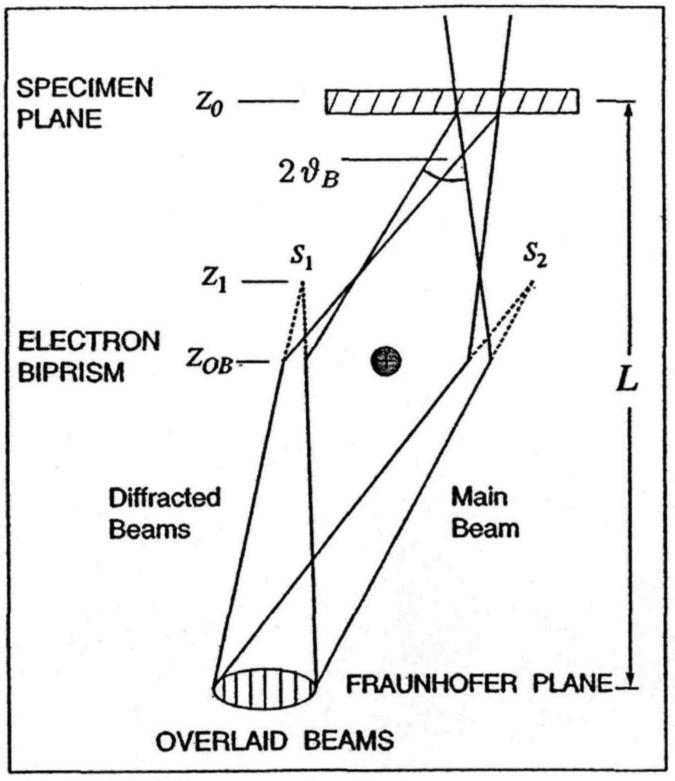
While working on the Tonomura Electron Wavefront Project, Exploratory Research for Advanced Technology (ERATO) for the Japan Science & Technology Corporation in the early 1990's, a method of electron holography referred to here as Diffracted Beam Holography (DBH) was invented [2]. Figure 1 shows a simplified schematic of the ray path of the electron beams. A primary beam from the electron gun is amplitude split by a crystal into a main beam and many diffracted beams where only one diffracted beam is shown here. An electron biprism is placed between any two beams having a positive potential, which deflects the two beams towards each other so they can interfere to form an interferogram on an observation plane (Figure 1b). The interferogram can be considered a hologram if a good reference beam can be established such as when a main beam is generated from a thin crystal so kinematic diffraction conditions exist and its structure factor can be considered zero.

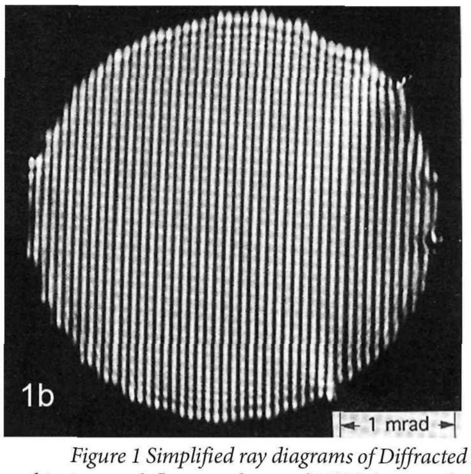
For DBH, the exact overlaid beams condition, as shown in Figure 1a, is comprised of five phase contributions, *i.e.*, the biprism ($\mathbf{K_B}$), the beams passing to the left ($\mathbf{g}_L$) and to the right ($\mathbf{g}_R$) of the biprism, which both suffer from the spherical aberrations of the focusing lenses (C_{SB}), and the two material phases ($\beta_{L,R}$) contained in the beams L, R passing to the left and right hand side of the biprism, respectively. Most often the goal is to measure the material's phase information, *i.e.*, $\beta_{L,R}$. This can be written as [2],

$$\phi = \begin{bmatrix} 4\pi r_{\mathbf{B}}\mathbf{K_B} \\ -C_{SB}(\mathbf{k}+\mathbf{g}_L)+C_{SB}(\mathbf{k}-\mathbf{g}_R) \\ +(\beta_L+\beta_R) \end{bmatrix}$$

where $r_{\mathbf{B}}$ is the radius of the biprism.

This method of electron holography has been able to measure the phase shifts around point defects, dislocations,





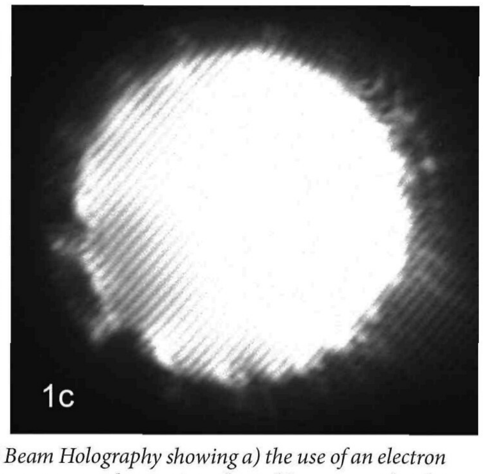
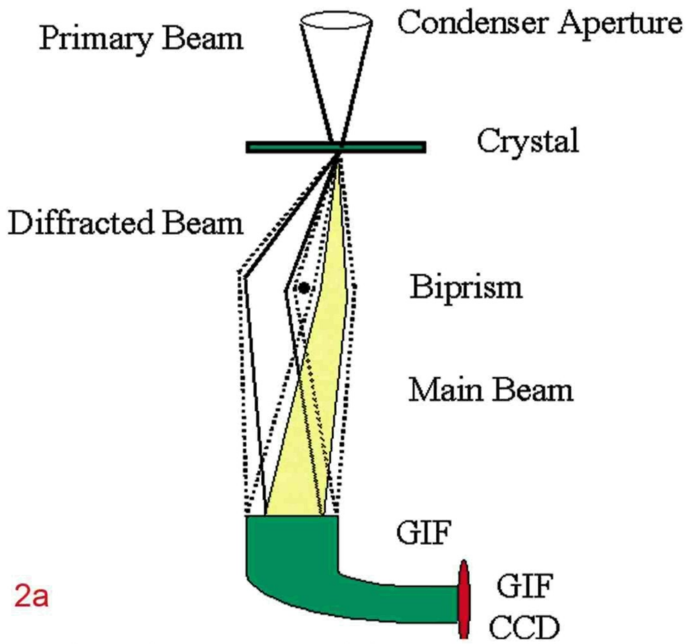


Figure 1 Simplified ray diagrams of Diffracted Beam Holography showing a) the use of an electron biprism to deflect two beams (DBH) for interference on an observation plane, b) an example of a hologram using the main beam and 111 diffracted beam of Gallium Arsenide, and c) an over-exposed hologram showing the fringes extending outside the boundaries set by an aperture, which suggested the interference of inelastically scattered electrons from the plasmons created inside the material by the electron beam irradiation



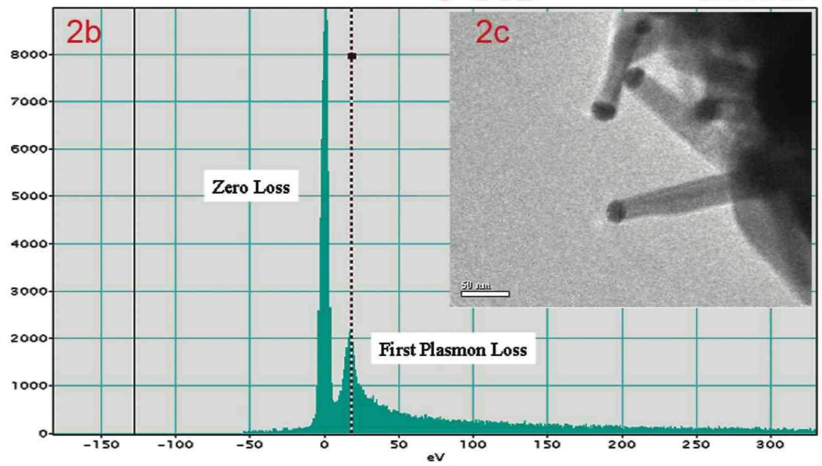


Figure 2 a) the configuration of energy-filtered electron holography showing an electron biprism deflecting the main and diffracted beams into the GIF, b) an electron energy loss spectrum of GaAs showing the zero-energy loss peak and first plasmon loss peak, which were used with a 5 eV window by the GIF to selectively interfere these electrons in the main and diffracted beams onto the GIF CCD, and c) the GaAs specimen consisting of tubes capped with gold having a diameter of 20-25 nm.

quantum well structures, *etc.* due to the strain and electrostatic fields existing in materials [3]. During the ERATO project, it was noticed that the electron interference in the holograms unexpectedly continued outside of the boundaries set by a beam limiting aperture (condenser aperture) (Figure 1c) and it was hypothesized that this interference was due to "energy-loss" or inelastically-scattered electrons, which may have interacted with either the bulk plasmons or phonons of the material, which would enable some of the inelastically-scattered electrons to deviate to the outside of the aperture [4]. At the time of the ERATO project, when this result was presented, the coherent interference of inelastically electrons was highly unexpected and doubted by those working in the electron holography field. Recent experiments though, which gave access to a field-emitting gun (FEG) STEM equipped with both an electron biprism and Gatan Imaging Filter (GIF), which is surprisingly a rare combination of FEG STEM accessories, have been able to verify the above-mentioned hypothesis, by combining the DBH method with an energy-filtering GIF to produce Energy-filtered Electron Holography.

DBH combined with a Gatan Imaging Filter (GIF) is shown in the schematic diagram of Figure 2a, where in this case the main beam is interfering with a diffracted beam. An electron biprism is placed in between the two beams and is used to deflect the beams towards each other for interference on the GIF CCD. The GIF uses an energy window to isolate the energy-loss electrons from the zero-energy loss electrons. Figure 2b shows an example of an electron energy loss spectrum taken from a GaAs specimen, which is shown in Figure 2c. In Figure 2b, two peaks are observed, one due to the zero-energy loss electrons and the other due to the first plasmon-loss electrons. Only one plasmon-loss peak is created by the very thin (20-25 nm) specimen. In the example holograms shown in Figure 3, which used the same GaAs specimen of Figure 2c, the main beam is interfered with the 111 beam. In Figure 3a, the zero-loss electrons from the two beams were interfered and all of the fringes were contained within the aperture. In Figure 3b, the plasmon-loss electrons from the two beams were interfered, which resulted in the formation of high-contrast fringes inside and outside of the aperture, although the edge of the aperture cannot be clearly resolved, and the contrast of the fringes leading away from the outside of the aperture become weak. The fringes inside the aperture had a high degree of coherence of ~0.3 as can be seen in the intensity spectrum shown in Figure 3c. The existence of the fringes outside of the aperture was only found for the holograms consisting of inelastically-scattered electrons, as previously hypothesized.

Complimentary measurements of the interference of inelastically-scattered electrons from bulk plasmons have been made using off-axis electron holography, *i.e.*, a wave-front-splitting method, combined with a GIF [5]. As well, another means to interfere inelastically-scattered electrons is by spatially resolved EELS [6]. Both of these methods differ from DBH primarily by observing the inelastically-scattered electrons at the specimen in-focus plane, where the contrast of the fringes in the plasmon-loss holograms are low, perhaps due to a low intensity of the beams being overlaid and multiple diffracted beams contributing to the image.

In conclusion, recent experiments combining DBH with a GIF have verified that inelastically-scattered electrons have sufficient coherence to interfere with themselves to form

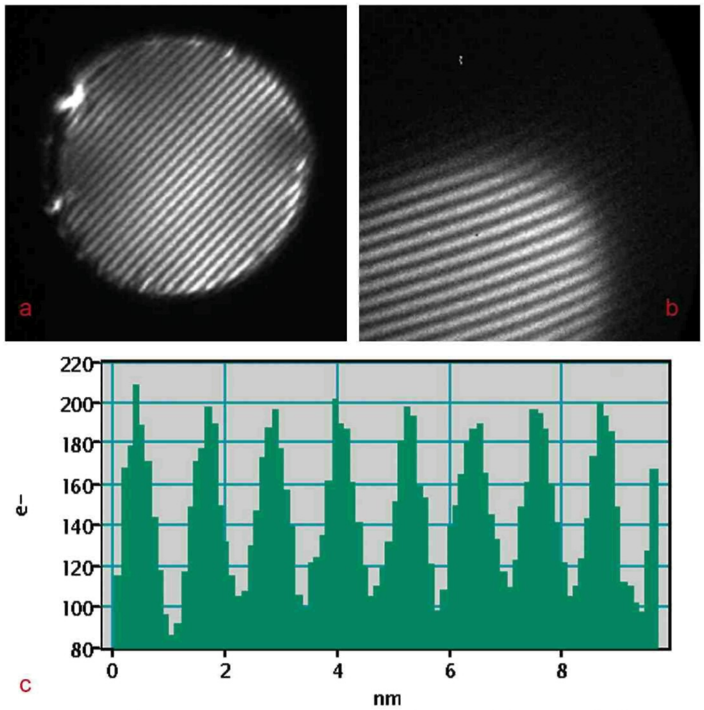


Figure 3 Holograms showing the main beam interfering with the 111 diffracted beam of GaAs where a) consists of zero-loss electrons, b) consists of first plasmon-loss electrons centered at 18 eV and having a 5 eV window and c) is its associate intensity profile.

fringes in the holograms both within and outside of the beam's aperture. In order to verify theoretical models of plasmon-loss electrons as a function of thickness, thicker specimens generating second and third plasmon-loss electrons need to be characterized by this method. These measurements are possible but dynamic diffraction effects would need to be considered. The fact that no fringes form around the aperture for the zero-loss electron holograms indicate that the phonon-scattered electrons, which scatter multiple times at low energies (<0.1 eV), either are not scattered coherently or their intensity is insufficient to be observed by the present experiments. Use of longer recording times, thicker specimens or lighter element specimens (*e.g.*, diamond) may resolve if phonon-scattered electrons also have a sufficient degree of coherence to form fringes. ■

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