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Practical procedure for retrieval of quantitative phase map for two-phase interface using the transport of intensity equation

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ABSTRACT

A practical procedure for retrieving quantitative phase distribution at the interface between a thin amorphous germanium (a-Ge) film and vacuum based on the transport of intensity equation is proposed. First, small regions were selected in transmission electron microscopy (TEM) images with three different focus settings in order to avoid phase modulation due to low frequency noise. Second, the selected TEM image and its three reflected images were combined for mirror-symmetry to meet the boundary requirements. However, in this symmetrization, extra phase modulation arose due to the discontinuous nature of Fresnel fringes at the boundaries among the four parts of the combined image. Third, a corrected phase map was obtained by subtracting a linear fit to the extra phase modulation. The phase shift for a thin a-Ge film was determined to be approximately 0.5 rad, indicating that the average inner potential was 18.3 V. The validity of the present phase retrieval is discussed using simple simulations.

1. Introduction

The potential distribution at two-phase interfaces is important for understanding the physical and chemical properties of devices. Recently, the local potential distribution at the interface between cathode (anode) materials and an electrolyte has been a key issue for improvements of lithium-ion batteries [1,2] since battery performance is related to local lithium diffusion at the interface, which depends on the local potential.

Transmission electron microscopy (TEM) is a powerful tool for determining not only the local structure, but also the local potential distribution, since the phase shift of the incident electron wave is proportional to the local potential. The phase shift can be measured using methods such as electron holography (EH) [3], focal series reconstruction methods [4], diffractive imaging [5] and the transport of intensity equation (TIE) [6–13]. Among these, EH, which reconstructs the phase using the interference between an object wave that passes through the specimen and a reference wave that passes through vacuum, is the most popular method. However, installation of a biprism into the TEM column is necessary in order to deal with interference, and a vacuum region for the reference wave is also essential. These requirements restrict the application of EH.

Phase information can also be retrieved by utilizing the principle of wave propagation, and the TIE is one such approach [6–13]. The TIE does not require any special microscope attachments or a vacuum region, and is thus considered to have a wider scope for applications compared to EH. The TIE has been used to determine the mean inner potential (MIP) of metallic nanoparticles [14–16], and it was found that the MIP increases as the particle diameter decreases to a few nanometers. Petersen et al. investigated the quantitative TIE-retrieved phase energetically from various aspects including precise determination of the defocus value and estimation of the modulation transfer function for a charge-coupled device (CCD) detector [17]. They also demonstrated a quantitative TIE-retrieved phase map for MgO nanocubes [18]. The TIE has also been used to observe helical spin order [19] and two-dimensional

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skyrmion crystal phases [20,21].

However, the phase distribution at the interface has been rarely investigated using the TIE method thus far. One of the reasons is the requirement of boundary conditions. The Fresnel fringes of an object, which contains the phase information, vary with defocus in a TEM image, and should not spread to the outside of the image area in order for phase retrieval. In the case of metallic nanoparticles adsorbed on a uniform substrate, a suitable region surrounding the nanoparticle can be selected to contain all Fresnel fringes of the particle. However, as for an interface, the Fresnel fringes in the vicinity of the interface are subject to be cut off by selecting a region, which causes artifact in the retrieved phase. This is because the discontinuous Fresnel fringes at the boundary are assumed to be periodic and continually connected with each other when carrying out Fourier transform (FFT) calculation. Volkov et al. proposed a mirror-symmetrization method to solve such problem [10]. If the original TEM image meet the intensity conservation law, which means the total intensity is maintained regardless of defocus, then the phase shift at the interface can be correctly retrieved. This has been demonstrated effective through simulation in their article.

Low-frequency noise has been shown to be amplified, which is also a problem for quantitative phase retrieval [11,12,17,22]. The phase change at a two-phase interface has been reconstructed by employing an image mask for noise subtraction [22]. However, these methods do not seem to be sufficiently practical.

In this study, we demonstrated that the phase distribution can be quantitatively retrieved for the interface between a thin amorphous germanium (a-Ge) film and vacuum using the TIE-FFT method. Using a selection of small regions in the TEM images was effective at removing low-frequency noise. In addition, mirror-symmetrization of the experimental TEM image and subtraction of the linear background was found to be necessary.

2. Experimental methods

TEM observations were conducted using a 50 pm resolution electron microscope (R005) equipped with a cold field emission gun and double spherical aberration-correctors, operated at an accelerating voltage of 300 kV [23]. In this study, TEM images were obtained with a spatial resolution of 0.84 nm/pixel. A through-focus image series was obtained from −10 μm (under-focus) to +10 μm (over-focus) in steps of 2 μm by changing the objective lens current. Under-focus usually has minus sign in this study. Aberration of the imaging lens was corrected below the fourth-order and the chromatic aberration was 1.65 mm. The TIE phase retrieval procedure was performed using the QPt (HREM Research Inc.) plug-in software [24] for Digital Micrograph (Gatan Inc.). In this procedure, three TEM images (under-, in- and over-focus) were aligned at the sub-pixel level.

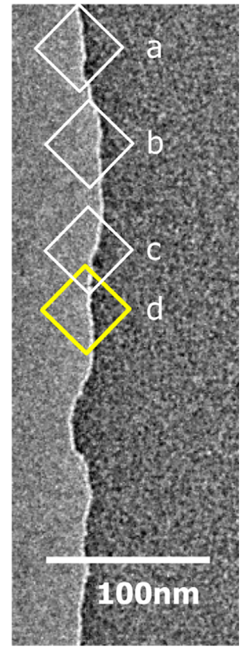

Fig. 1. A typical TEM image of the interface between a thin a-Ge film and a vacuum taken at under-focus 8 μm. Four TIE-retrieved phase maps were obtained at the small regions denoted by (a – d).

For the analysis, the MIP was calculated using

$$\phi(x) = C_E V_0 t(x) \tag{1}$$

where C_E is a constant that is related to the electron energy (C_E=6.523 × 10^{-6} rad V^{-1} nm^{-1} at an acceleration voltage of 300 kV), V_0 is the MIP (assuming that the MIP is constant) and $t(x)$ is the thickness distribution. Using Eq. (1), V_0 could be calculated when $t(x)$ and the phase shift $\varphi(x)$ were experimentally obtained.

The thin a-Ge film used in this study was a commercial product. As a simple case, the film/vacuum interface was considered to be better than a solid/solid interface to clarify the problem that whether the potential distribution at the interface could be estimated quantitatively by TIE method. The present phase retrieval method is also thought to be useful for a solid/solid interface under certain conditions.

For TEM observations, the current density needs to be as low as possible in order to suppress irradiation damage; however a certain amount of current density is necessary in order to improve the signal-to-noise ratio. Therefore, the optimal current density was determined by the balance between these two requirements.

The thickness of the thin a-Ge film was estimated from electron energy-loss spectroscopy (EELS) measurements. The EELS spectrum was obtained with an aperture size of 54 mrad, an exposure time of 0.05 s and an energy dispersion of 0.1 eV/channel. The inelastic mean free path λ, was calculated to be 153 nm using the expression given in