

Atomic Resolved Phase Map of MoS₂ Monolayer Sheet Retrieved by Spherical Aberration Corrected Transport of Intensity Equation

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An atomic sheet like graphene has attracted much interest for its unique physical and chemical properties. Using a spherical aberration corrected microscope, not only atomic defects [1] but also chemical bonding at the edge [2] have been measured. We think that retrieving a phase map, which represents the potential map, is also important in order to understand chemical reaction or charge distribution. Transport of intensity equation (TIE) is a convenient method for retrieving the phase map, since it does not require any special microscope attachments or a vacuum region. In this study, we retrieved atomic resolved the phase map of MoS₂ monolayer sheet by TIE. In a TIE phase map, the defocus difference between two TEM images should be reduced in order to improve spatial resolution [3]. Aberration correction electron microscopy is thought to solve such a problem, because it has a high spatial resolution and show visible contrast variation by changing the sign or amount of defocus around in-focus condition.

By using an aberration correction electron microscope, R005, having 50pm resolution, a MoS₂ monolayer sheet was observed at an accelerating voltage of 300kV. The current density was kept as low as possible during observation. Through-focus series of high-resolution TEM images were obtained at 2 nm step in the range from -20 nm (under-focus) to 20 nm (over-focus). Atomic resolved phase maps were retrieved from two TEM images of -8 nm and 8 nm defocuses as shown in Fig. 1(a) and (b), as an example.

An atomic-resolved TIE phase map was obtained by applying high pass filter to the original map as shown in Fig.1 (c), to remove low frequency noise. In Fig.1 (c), molybdenum (Mo) and sulfur (S) atomic columns seemed to correspond to the right and left peaks of the dumbbell curve, since the right peak is of slightly higher contrast than the left one. In order to confirm this statistically, 25 different phase profiles of the dumbbells were investigated. During analysis, the background was subtracted from the raw phase profile, since the background was modulated due to low frequency noise and so on. After subtraction, the left-side and right-side peaks of the dumbbell and the valley between both peaks were measured. The phase differences between the left-side peak and valley, and between the right-side peak and valley were obtained to be 0.045 and 0.058 radian on average, respectively.

Phase map of MoS₂ monolayer sheet were obtained by multi-slice simulation. It showed sharp peaks corresponding to Mo (0.435 rad.) and 2S, consisting of two sulfur, (0.459 rad.) atomic columns. But, actually, spatial resolution is reduced depending on defocus difference in the TIE phase map. Several phase maps were obtained by applying low pass filter of 20, 13.33, 10, 6.67, 5 nm⁻¹ to the original phase map. And, Mo column, 2S column and the valley between them were measured. The phase differences between Mo (2S) column and valley were estimated to be 0.21 (0.20) (rad.), 0.12 (0.10), 0.062 (0.05), 0.015 (0.01) and 0.002 (0), respectively, for low pass filter of 20, 13.33, 10, 6.67, 5 nm⁻¹. Since the experimental phase differences were 0.045 and 0.058 radian, we determined that the left-side peak correspond to 2S column and the right-side, to Mo column. This determination is in agreement with the

phase map of Fig.1 (c), showing the right peak is slightly higher contrast than the left one. And also, the spatial resolution was determined to be about 0.1 nm in the present phase map.

In conclusion, we demonstrated that atomic resolved phase map of MoS₂ monolayer sheet was retrieved by transport of intensity equation (TIE) using spherical aberration corrected transmission electron microscopy. The phase shifts by Mo and S atomic potential was confirmed to be measured quantitatively.

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References:

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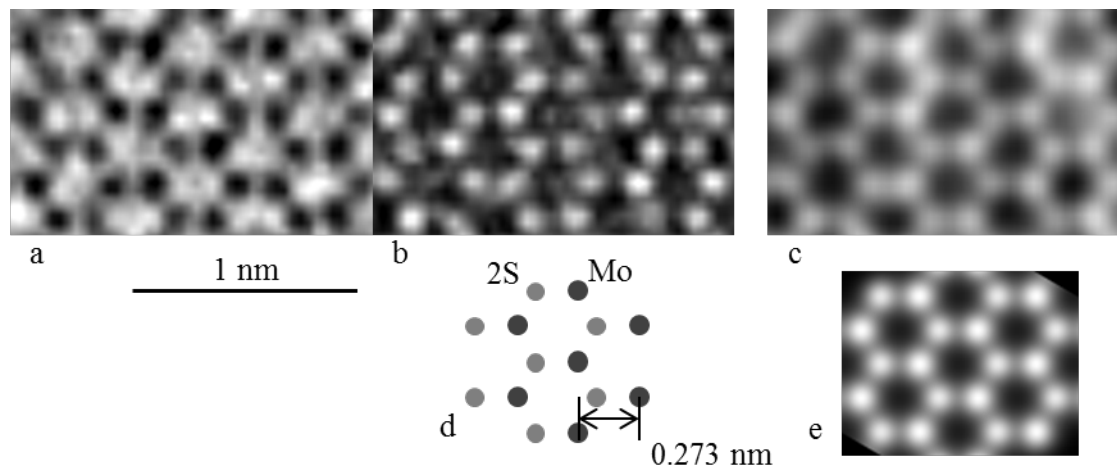


Figure 1. A TEM image of MoS₂ atomic sheet (a) at under-defocus of 8 nm and (b) at over-defocus of 8 nm. (c) A TIE retrieved phase map, (d) Structure model of MoS₂ atomic sheet and (e) simulated phase map, which was blurred by low pass filter.