

Quantitative transmission electron microscopy of two-dimensional transition metal dichalcogenides

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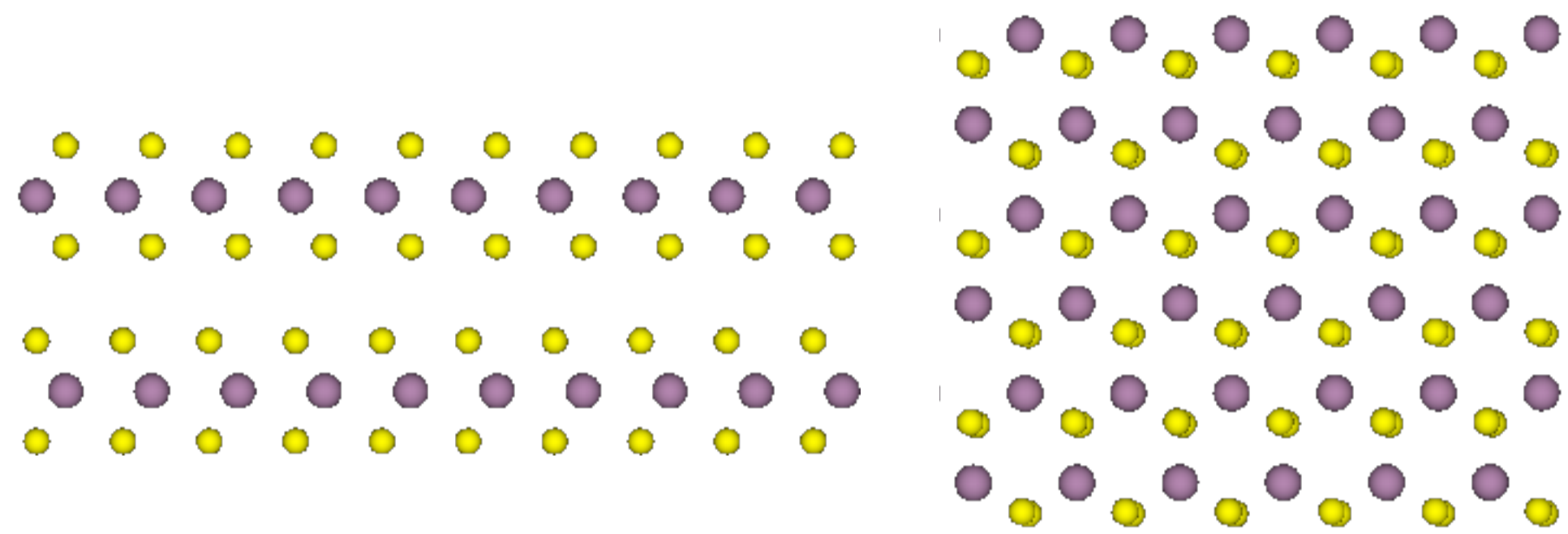
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Introduction

- Van der Waals bound layered structures.
- MX₂ (M: Mo, W; X: S, Se).
- Promising properties for spin-(opto)electronics: LEDs, solar cells, spintronic devices.



- There is a pressing need to obtain a full understanding of the local microstructural, chemical and functional properties of such layers with atomic spatial resolution using advanced TEM techniques.

Off-axis electron holography

- The TEM technique of off-axis electron holography provides access to the phase shift of the electron wave that has passed through a thin specimen:

$$\phi(x) = C_E \int V(x, z) dz - \frac{e}{\hbar} \iint B_{\perp}(x, z) dx dz \quad z: \text{direction of the incident electron beam}$$

- The phase shift is sensitive to the electrostatic potential V and the magnetic induction B . In the absence of magnetic and long-range electrostatic fields and assuming a uniform specimen structure and composition in the z direction:

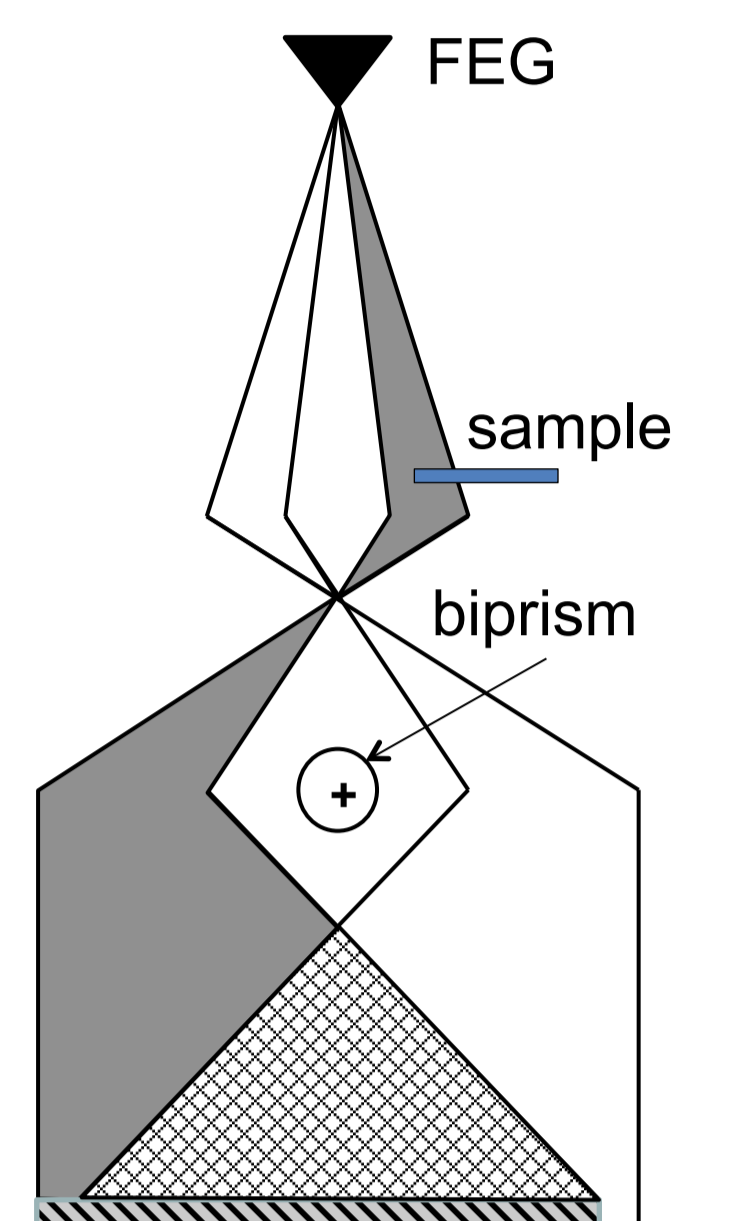
$$\phi(x) = C_E V_0(x) t(x) \quad C_E: \text{interaction constant; } V_0: \text{mean inner potential; } t: \text{specimen thickness}$$

- Values of V_0 can be calculated, to a first approximation, by regarding the specimen as an array of neutral free atoms:

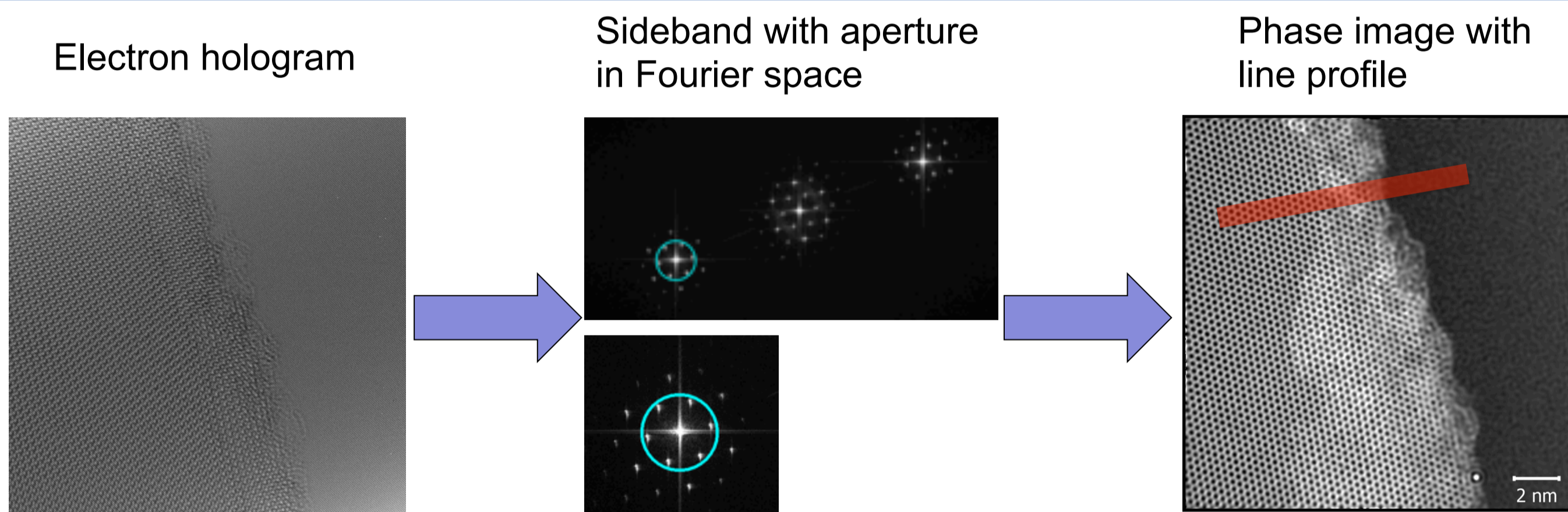
$$V_0 = \frac{\hbar^2}{2\pi m_e e \Omega} \sum_{\Omega} f_{el}(0) \quad f_{el}(0): \text{electron scattering factor; } \Omega: \text{unit cell volume}$$

- For WSe₂, the predicted phase shift per monolayer is:

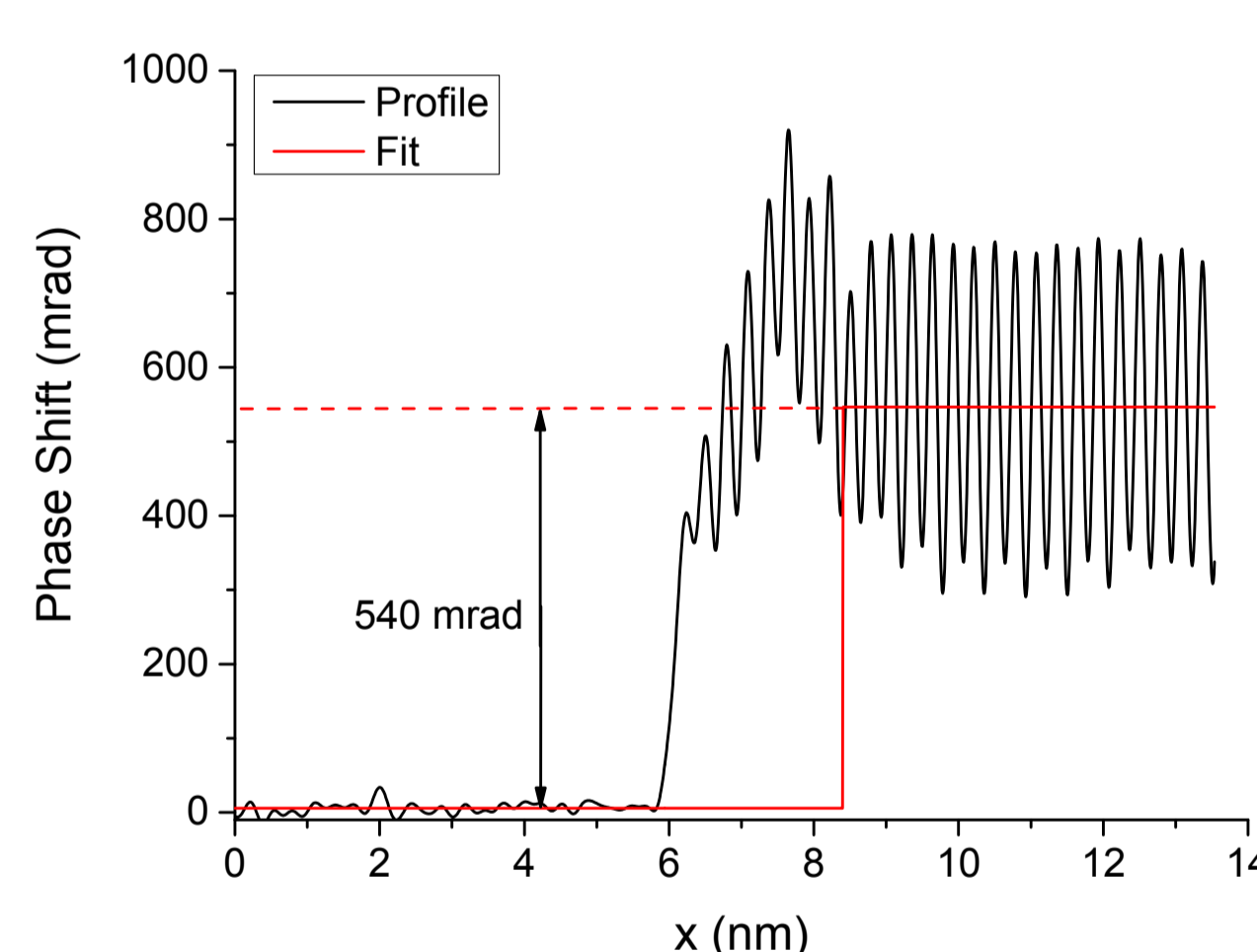
60 kV	80 kV
158 mrad	141 mrad



Experimental measurement of phase shift

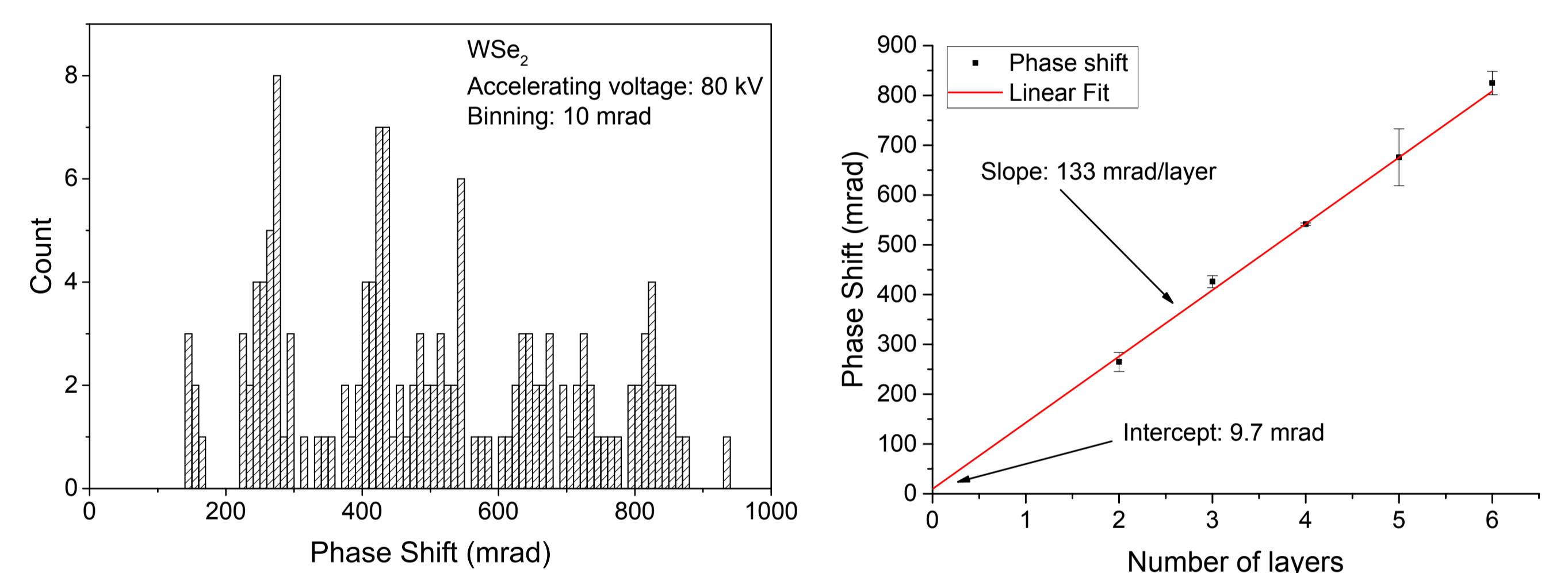


- Electron hologram recorded at a microscope accelerating voltage of 80 kV.
- Phase image reconstructed using a 15 mrad aperture to limit the effects of aberrations on the phase shift.
- Vacuum area flattened before extracting the line profile.
- The edge of the specimen has a non-uniform thickness.



Quantitative interpretation of phase images

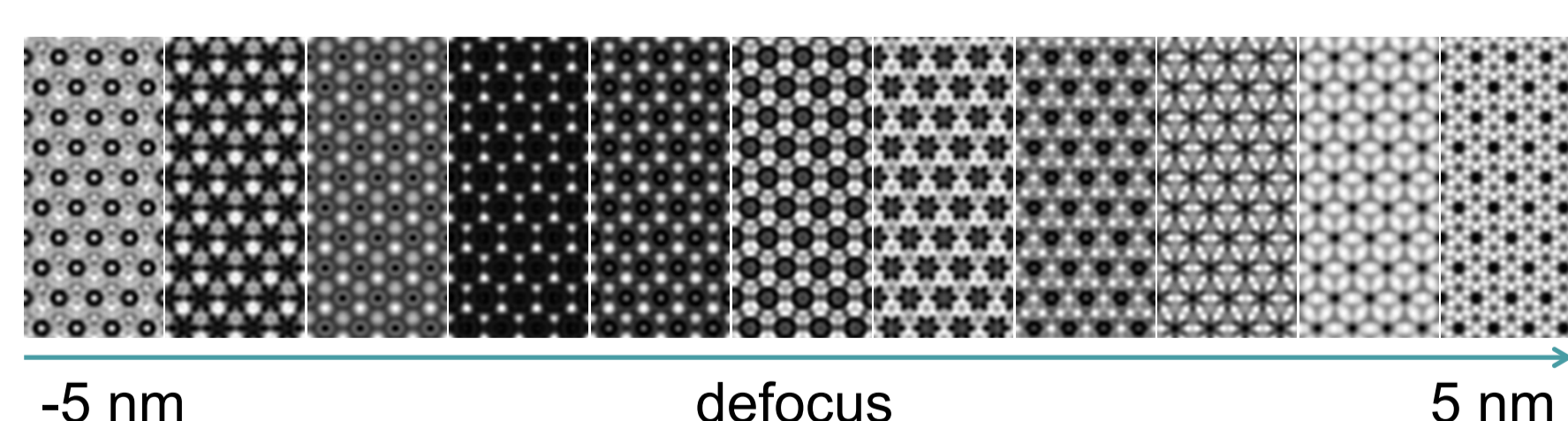
- Histogram of phase shifts measured from more than 70 line profiles:



- The experimental phase shift per monolayer (133 mrad) is lower than the predicted value (141 mrad) due to the effects of bonding on V_0 .
- The intercept of 9.7 mrad at 0 monolayers may originate from surface effects, such as:
 - The presence of a step in electrostatic potential induced by surface dipole layers (e.g. H₂O molecules).
 - The presence of ~0.1 nm (on average) of amorphous C.

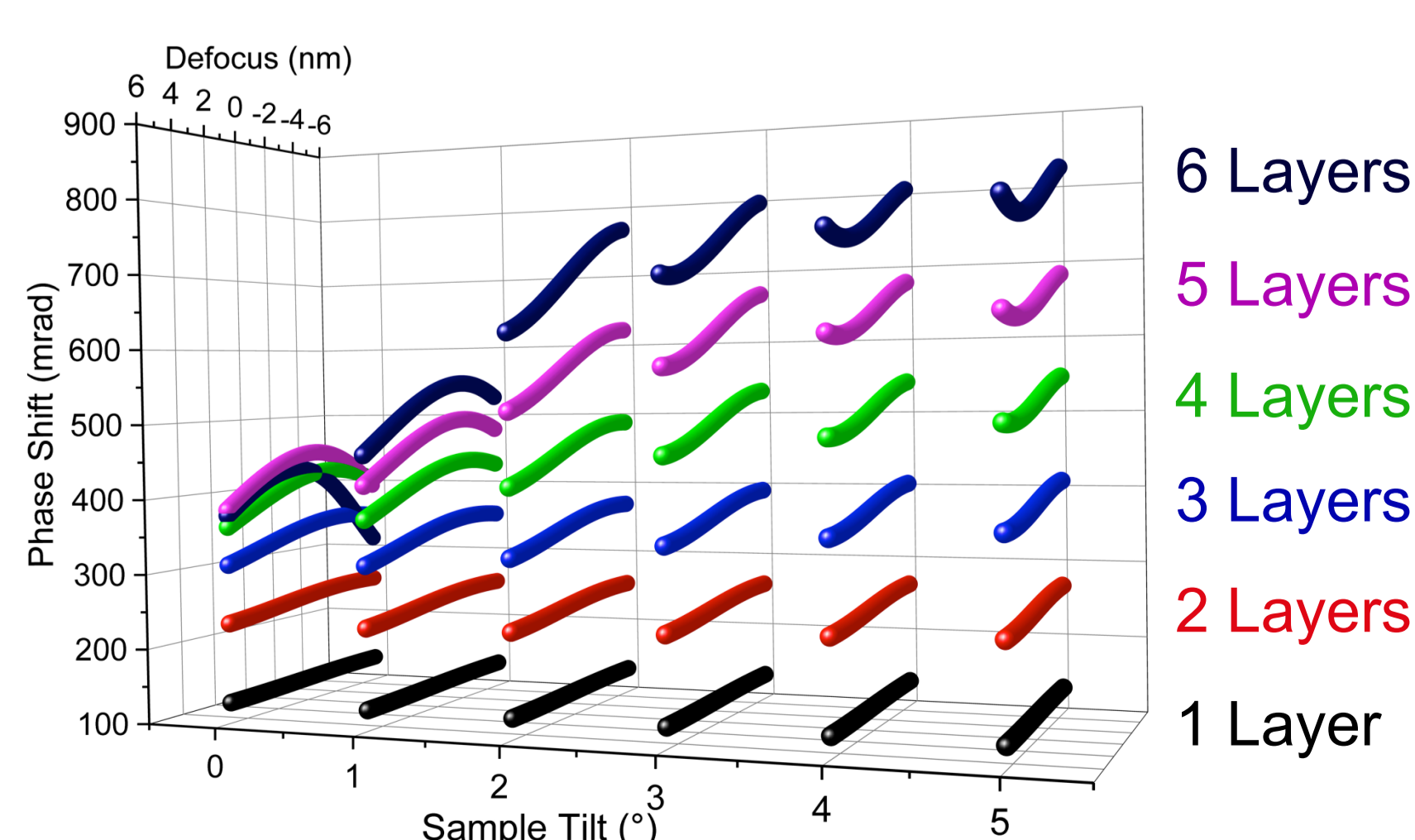
Comparison with simulated high-resolution phase images

- High-resolution phase images of monolayer WSe₂ simulated using Dr. Probe software:



- Average phase shift:
- Strong dependence on defocus
 - Non-linear for increasing number of layers

- The simulations show that defocus and other aberrations play a major role in determining the average phase shift measured from high-resolution phase images



Summary

- Experiments show a phase shift of 133 mrad per monolayer of WSe₂ and an extrapolated phase shift of 9.7 mrad at 0 monolayers.
- Simulations show a strong dependence of average phase shift on defocus and other aberrations.
- Small specimen tilts are needed to avoid dynamical effects and to obtain linear behaviour between phase shift and number of layers.

References

- 1) R.E. Dunin-Borkowski et al., in *Encyclopedia of Nanoscience and Nanotechnology, Chapter 3*, pp. 41-100, 2004.
- 2) Dr Probe software: <http://www.er-c.org/barthel/drprobe/>
- 3) D.K. Saldin and J.C.H. Spence, *Ultramicroscopy*, 55(4):397 - 406, 1994.

Acknowledgments

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