

Contamination and charging of amorphous thin films in a transmission electron microscope

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Introduction

Contamination and charging is a limiting factor in transmission electron microscopy (TEM) and can lead to artifacts especially in scanning (S)TEM, where the electron beam is focused on the sample^[1,2]. On the other hand it is exploited, e.g., in hole-free phase plate (HFPP) TEM^[3,4].

Contamination: Deposition of adsorbed molecules on the specimen surface by illuminating electrons.

Charging: Generation of charges on/in the specimen leading to unwanted electrostatic potentials.

Both effects were studied using a special electron-optical setup of a Hitachi HF3300 (Figure 1). The setup allows the acquisition of electron energy loss spectroscopy (EELS) under focused electron-beam illumination and HFPP image series.

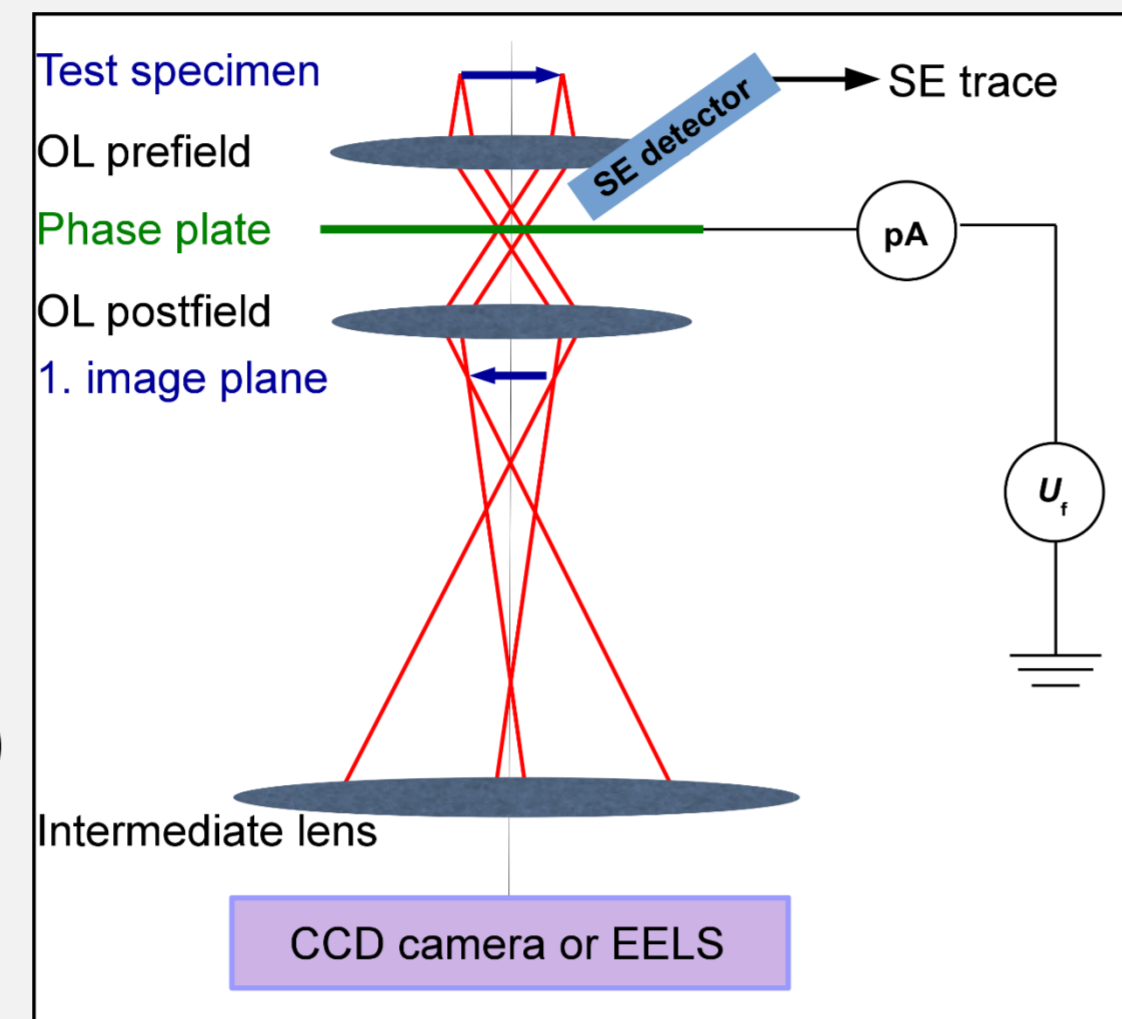


Fig. 1: Electron-optical setup to study contamination and charging of thin films under focused electron-beam illumination.

EELS → Information on thickness changes by low-loss spectra.

HFPP → Information on charging by power spectra analysis.

The following thin films were investigated:

- PVD/EB aC: 10/12 nm amorphous C (aC) by e⁻-beam physical vapor deposition (PVD) in a Lesker PVD75/Leica MED020.
- Thread aC: 7 nm aC thin film by C-Thread evaporation.
- PCS: 9 nm metallic glass alloy Pd_{77.5}Cu₆Si_{16.5} by sputter deposition.
- SiO₂: 10 nm SiO₂ by e⁻-beam (EB) PVD deposition.

Contamination and its inhibition

Illumination of the thin film shortly after insertion in the microscope leads to a strong increase of the relative thickness t/λ (Figure 2).

Contamination originates from the deposition of hydrocarbon contaminants on the thin-film surface induced by the electron beam with surface diffusion of adsorbed contaminants playing a major role (Figure 3).

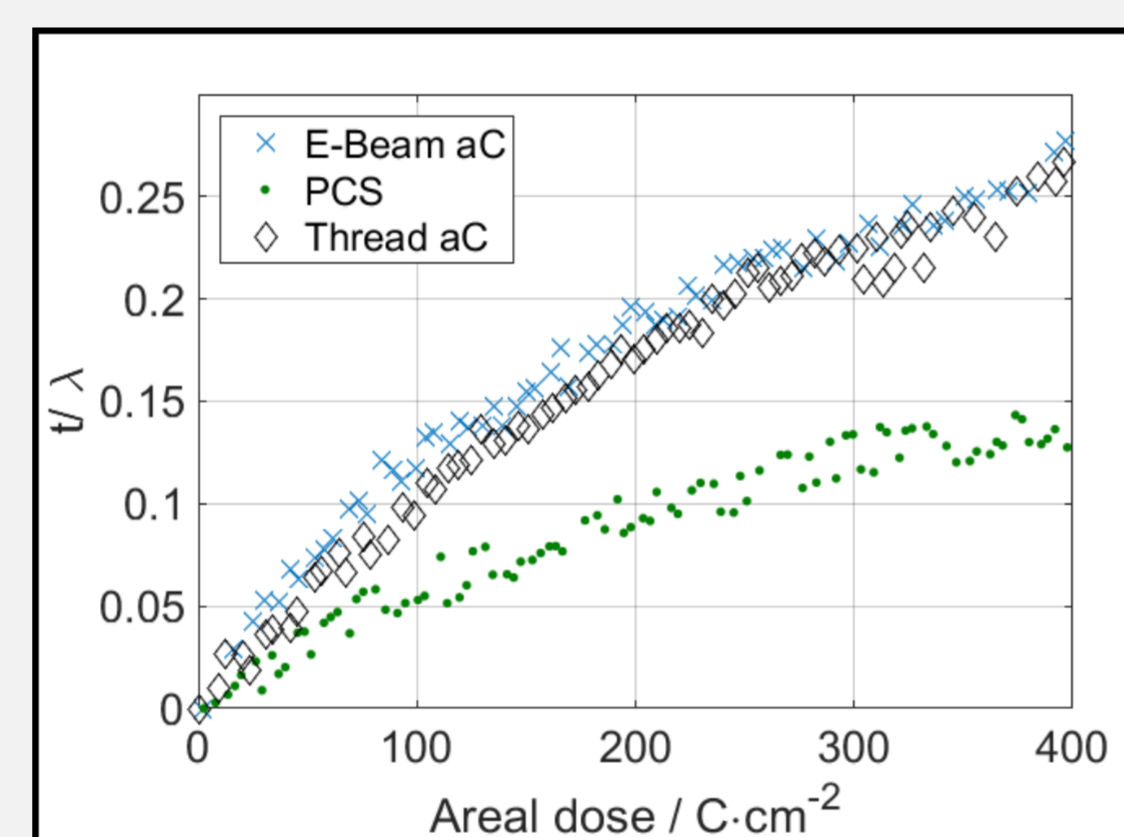


Fig. 2: Exemplary t/λ evolutions for the EB aC (blue), the PCS (green) and the Thread aC (black) thin film

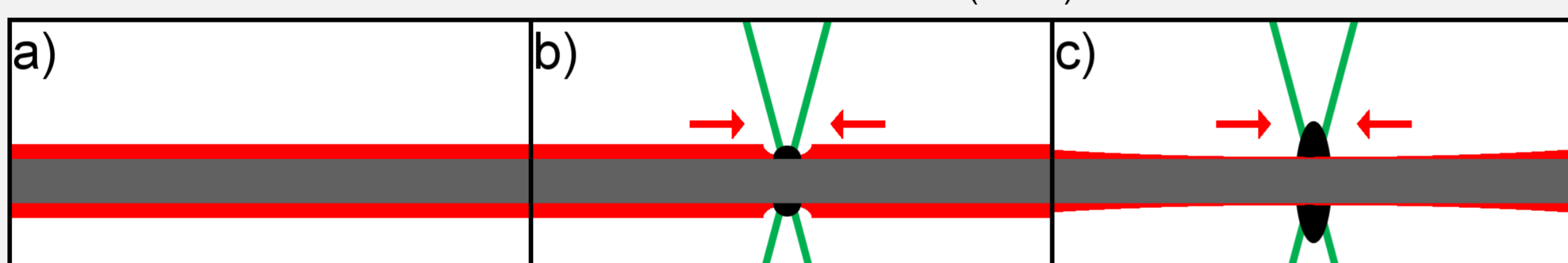


Fig. 3: Schematic description of contamination: adsorbed molecules (red) are deposited by the electron beam (green) to form a contamination layer (black). Molecules available for deposition are supplied by surface diffusion.

A detailed analysis of the acquired EEL spectra shows that the deposited contamination is similar to graphitic carbon (Figure 4) which is supported by a measurement of the sp_2 -fraction of the contamination layer^[5].

The measured t/λ can be transferred to a phase shift $\varphi = C_E t V_{MIP}$ if values for V_{MIP} (9 ± 1 V) and λ (150 ± 15 nm) of the deposited contamination layer are assumed. Figure 5 shows a comparison of the expected φ and φ determined by HFPP imaging. The curves agree well suggesting that no additional charging is present in the contamination layer.

Contamination can be inhibited by, e.g., UV cleaning or heating (Figure 6).

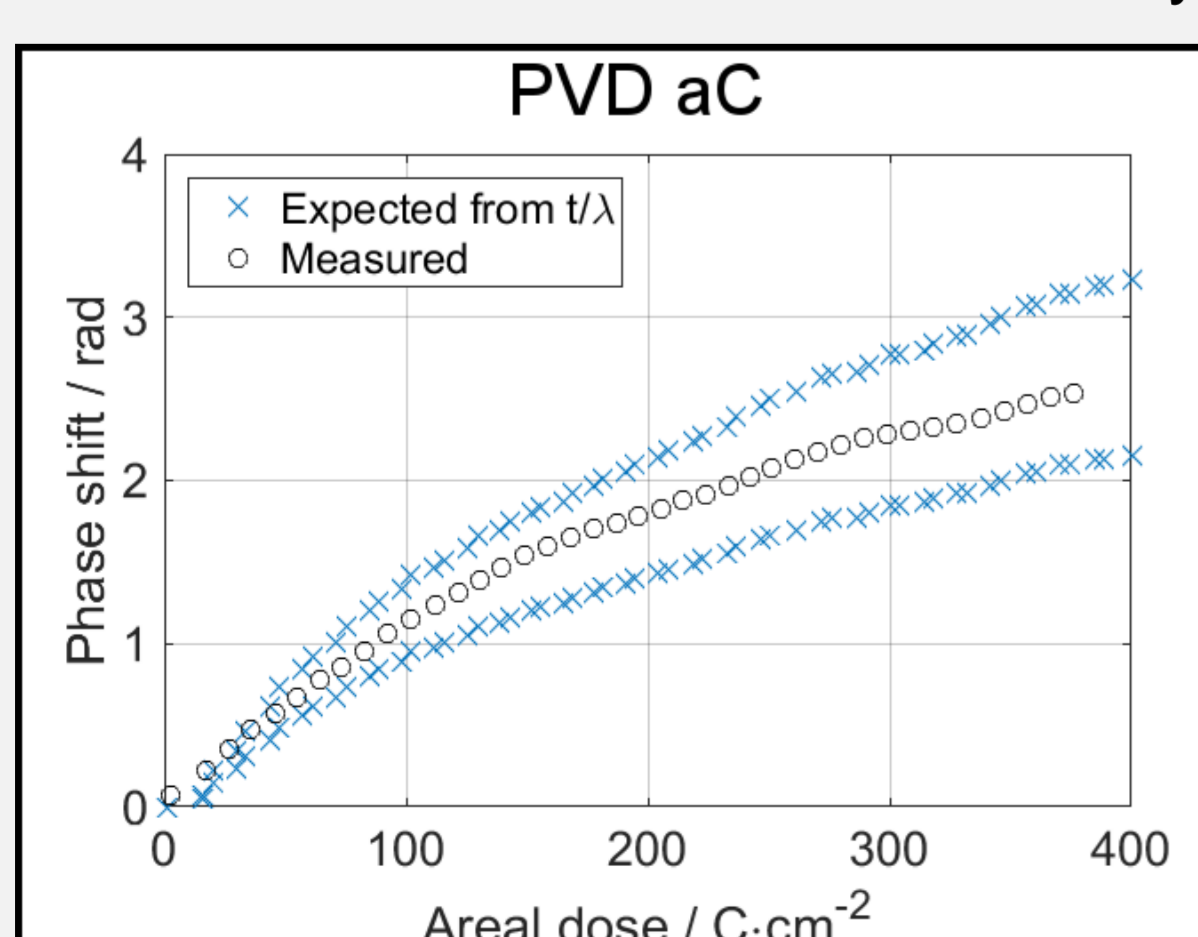


Fig. 5: Comparison between measured PS and the PS expected from the t/λ evolution.

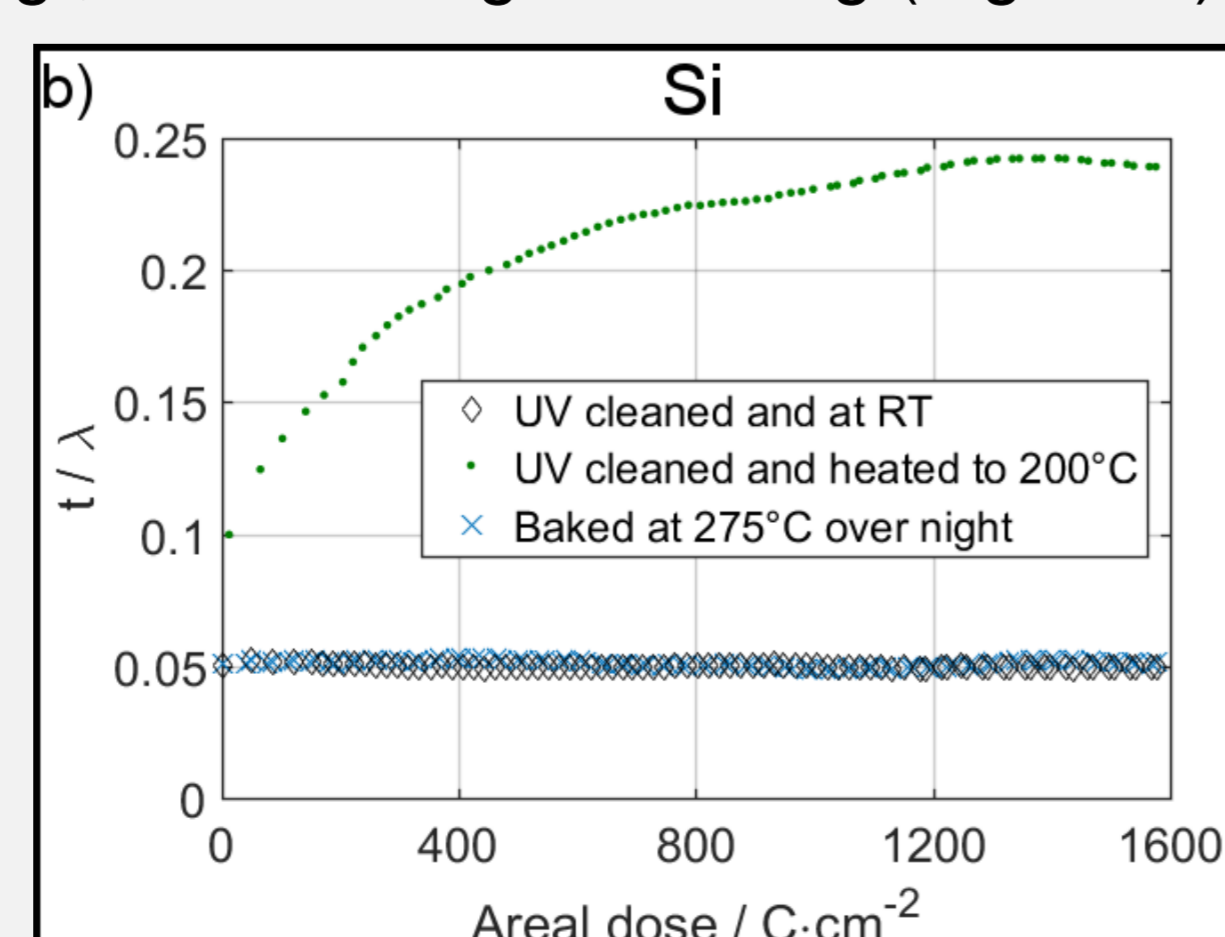


Fig. 6: t/λ evolutions after UV cleaning and subsequent heating of the Si thin film.

Charging and a possible explanation

Heating the Thread aC thin film to 275°C overnight leads to a thin film free of contamination and charging (Figure 7).

EB aC, PCS and SiO₂ thin films show negative PS if they are cleaned by either in-situ overnight heating (EB aC) or UV light (Figure 8). The PS cannot be explained solely by the slightly decreasing t/λ (Figure 9) suggesting additional negative charging. The decreasing thickness indicates that material is removed by the electron beam via electron-stimulated desorption (ESD).

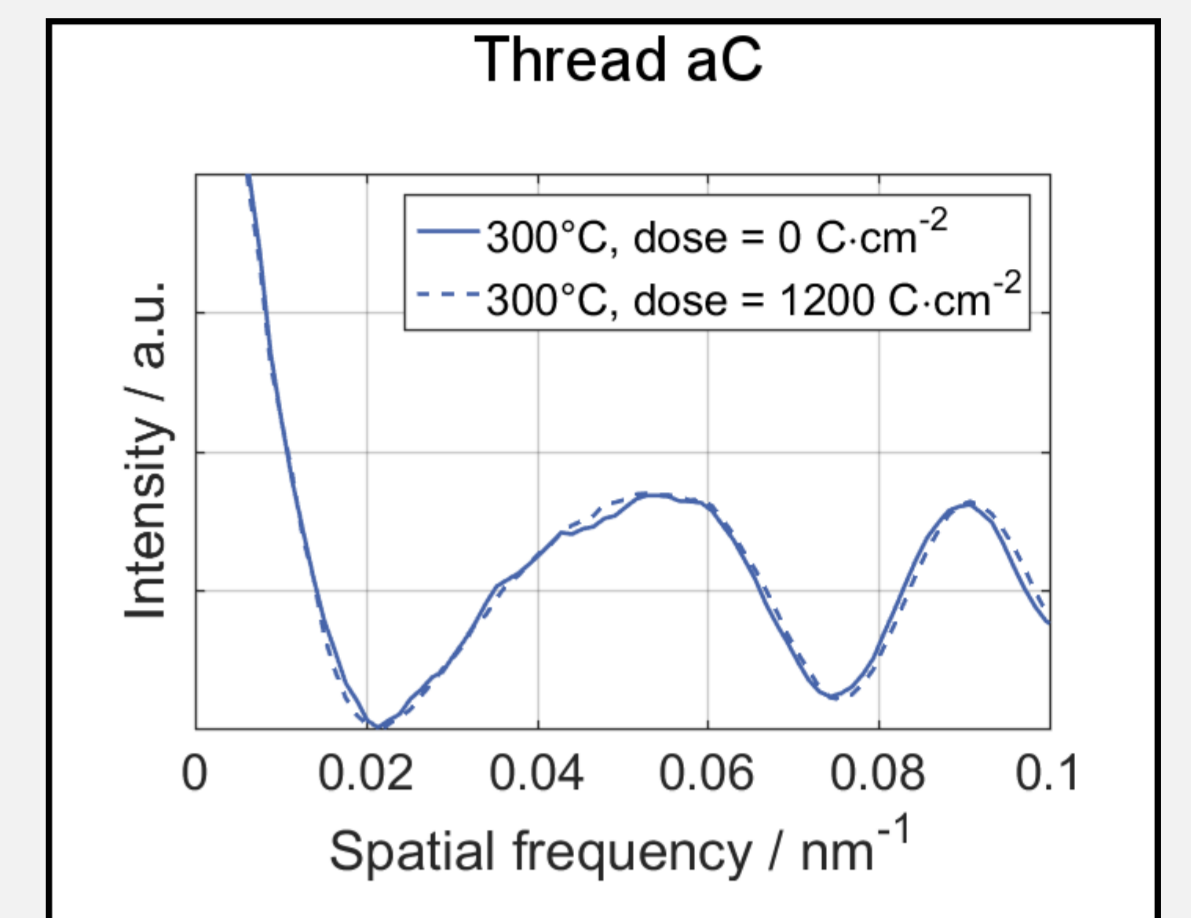


Fig. 7: Identical PCTFs before and after a HFPP-series show no charging.

PS measurement by HFPP imaging for cleaned thin films. EB aC, SiO₂ and PCS show negative charging. EB aC is heated in-situ at 275°C overnight and PCS and SiO₂ thin films are ex-situ UV cleaned to avoid contamination.

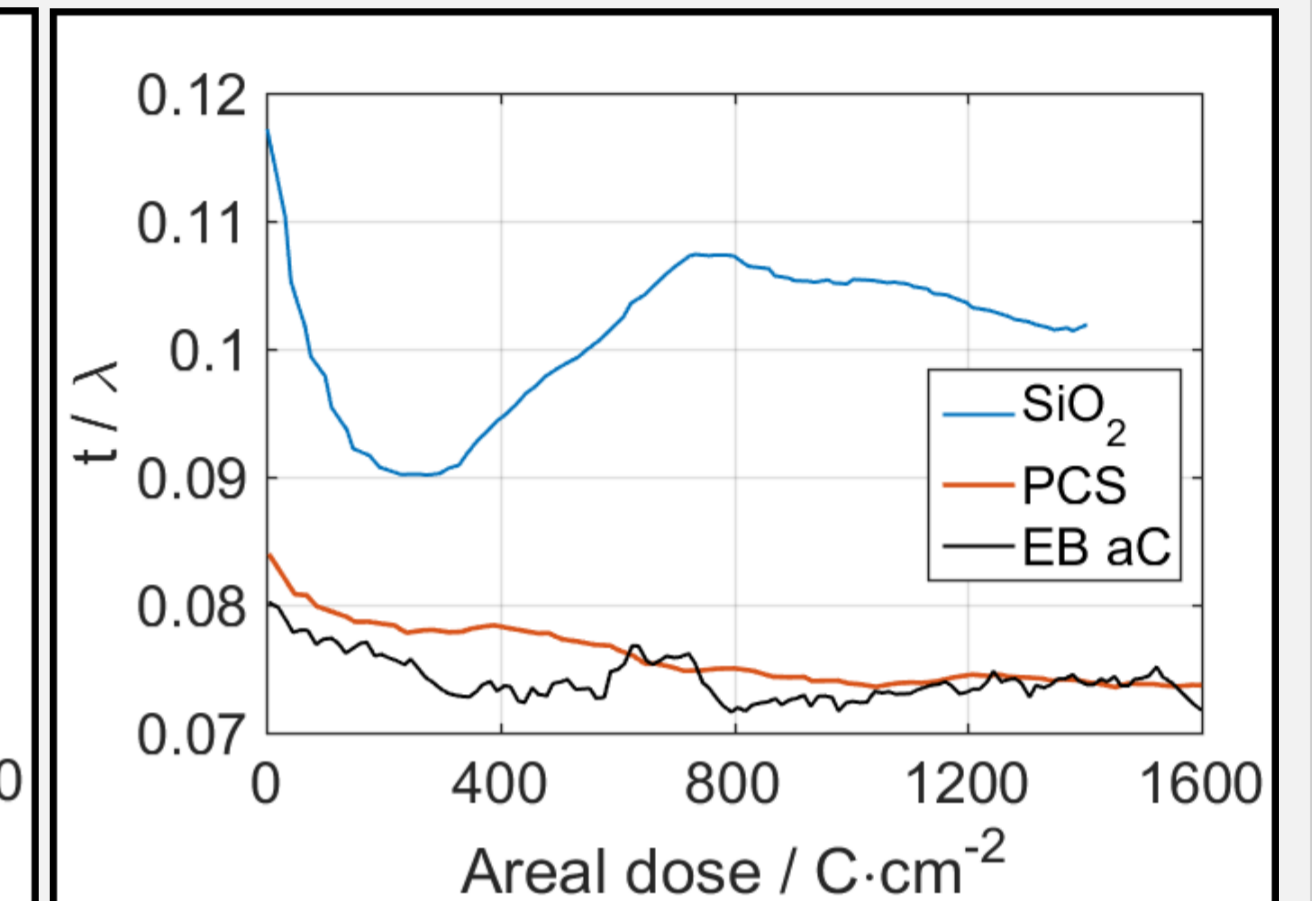
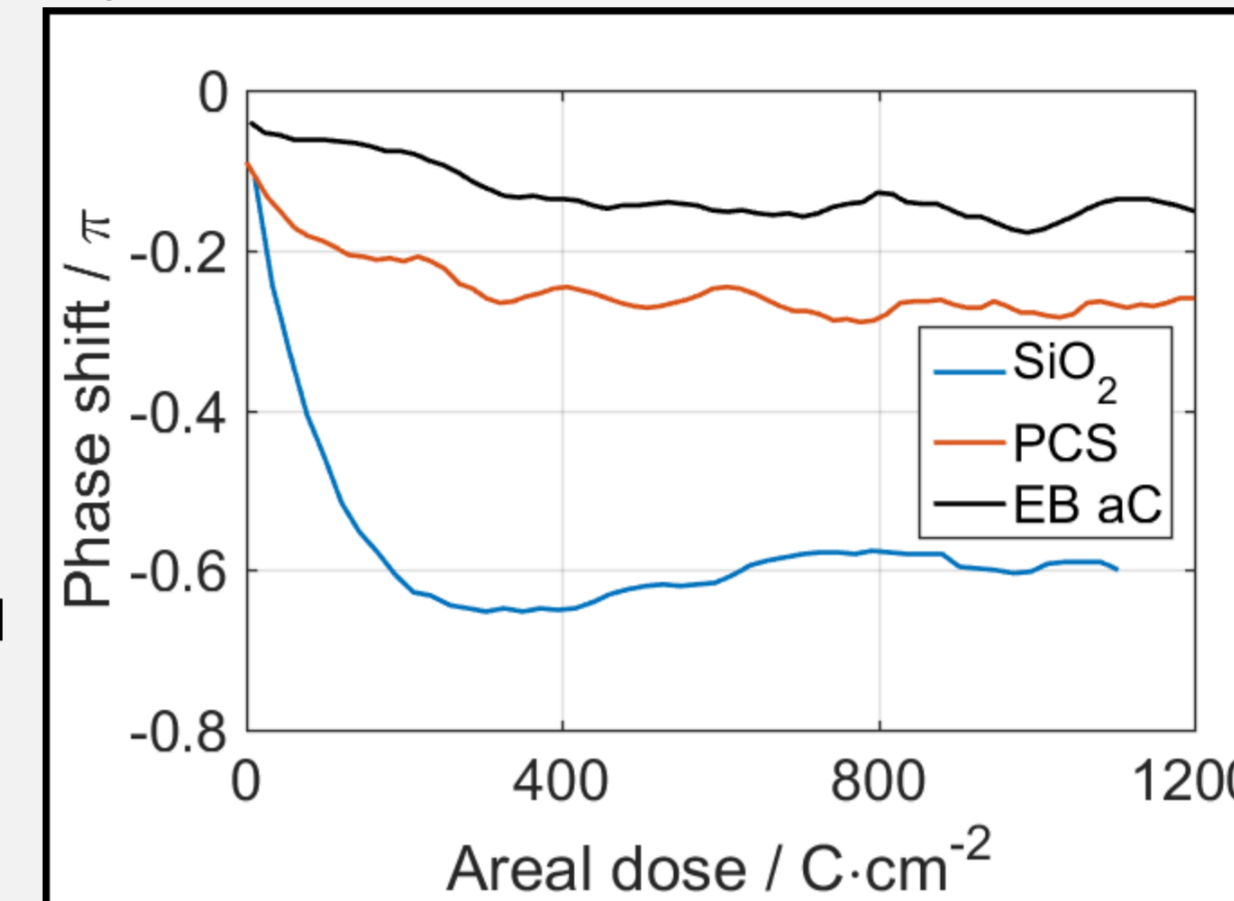


Fig. 9: t/λ measurements corresponding to Fig. 8 show a decreasing t/λ of the thin films.

Studies of the work function ϕ of metals revealed that adsorbed H₂O molecules can lead to a decrease of ϕ ^[6,7]. Adsorbed H₂O molecules can be interpreted as a surface dipole with the positive (H)

side facing to vacuum^[7] (Figure 10b) which causes a potential step δV . We propose that the observed negative charging of aC thin films stems from an ESD-induced change of ϕ in the irradiated area (Figure 10).

The induced PS in this model is given by:

$$\varphi = C_E R \delta V$$

Numerous thin-film properties and experimental conditions will affect φ :

- Temperature
- Surface roughness
- Porosity
- ϕ
- Conductivity
- Vacuum
- Beam current density

The described effect is a possible explanation for the functionality of the HFPP^[3] or Volta PP^[4].

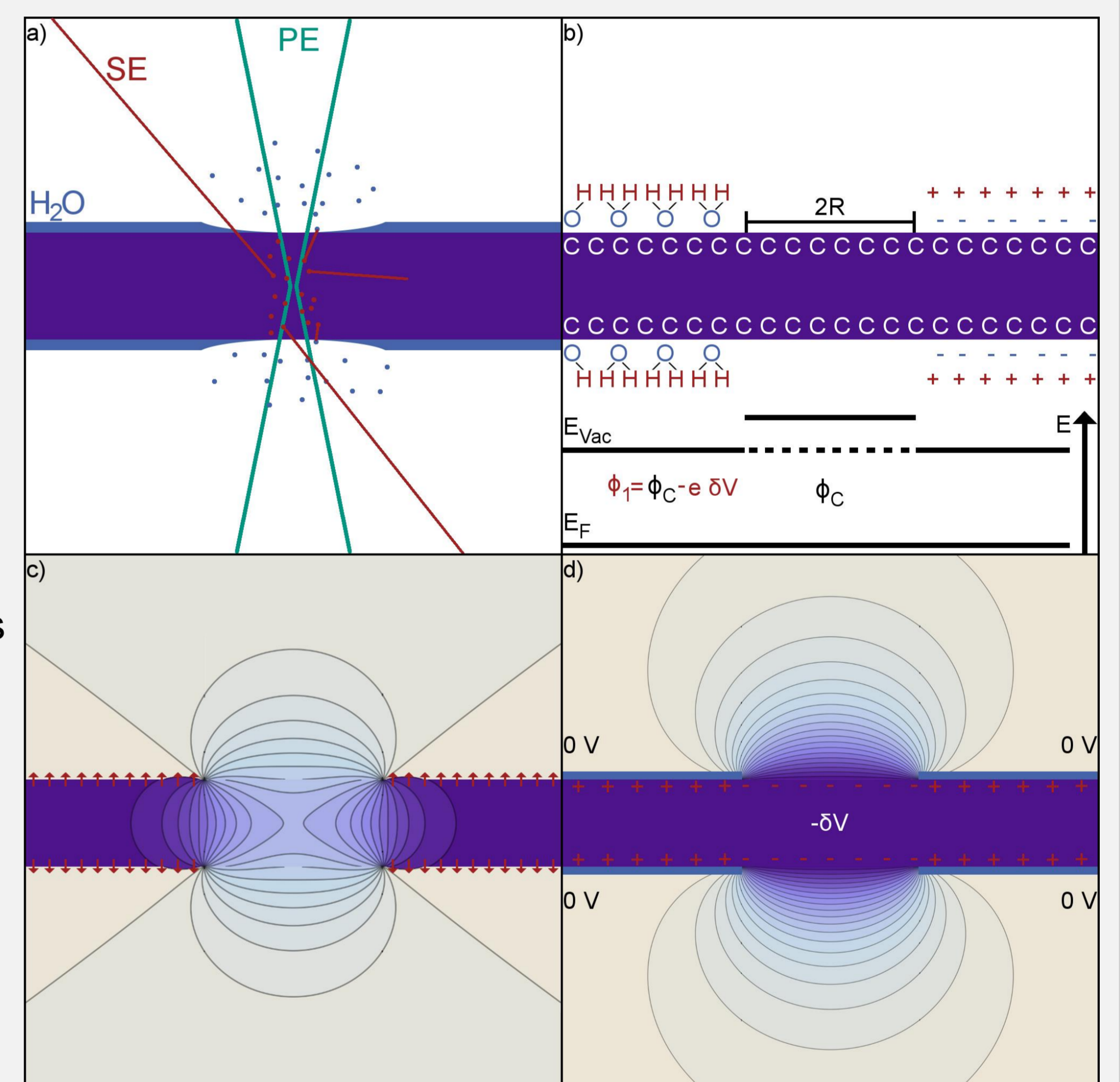


Fig. 10: Schematic illustration of the formation of negative charge induced by ESD and a subsequent ϕ change. (a) PEs generate SEs which induce ESD of H₂O. (b) The desorption of H₂O causes a ϕ step of $-eV$ in a circular area ($2R$). The electrostatic potential of the interrupted H₂O layer is displayed without (c) and with (d) thin film. In this model the induced phase shift is proportional to R and $-eV$.

Summary

- Contamination formed under intense focused electron-beam illumination is similar to graphitic carbon and does not charge^[5].
- Several methods are suitable for the complete or partial inhibition of contamination^[5].
- Contamination-free thin films showed no or small negative phase shift under electron-beam illumination which can be attributed to negative charging of the irradiated area.
- The origin of the charge is explained by electron-stimulated desorption of adsorbed H₂O-molecules and a subsequent change of the work function. The resulting electrostatic potential causes the observed negative phase shift.
- The described charging phenomenon is dependent on numerous thin-film parameters and experimental conditions making variations between different thin films and microscopes likely.

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