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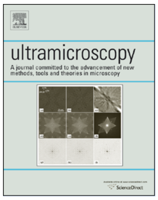

On the optical stability of high-resolution transmission electron microscopes

J. Barthel [a,c], A. Thust [b,c,*]

[a] Central Facility for Electron Microscopy (GFE), Aachen University (RWTH), Ahornstr. 55, 52074 Aachen, Germany
[b] Peter Grünberg Institute (PGI), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany
[c] Ernst Ruska-Centre for Microscopy and Spectroscopy with Electrons (ER-C), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany

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ABSTRACT

In the recent two decades the technique of high-resolution transmission electron microscopy experienced an unprecedented progress through the introduction of hardware aberration correctors and by the improvement of the achievable resolution to the sub-Ångström level. The important aspect that aberration correction at a given resolution requires also a well defined amount of optical stability has received little attention so far. Therefore we investigate the qualification of a variety of high-resolution electron microscopes to maintain an aberration corrected optical state in terms of an optical lifetime. We develop a comprehensive statistical framework for the estimation of the optical lifetime and find remarkably low values between tens of seconds and a couple of minutes. Probability curves are introduced, which inform the operator about the chance to work still in the fully aberration corrected state.

1. Introduction

The introduction of hardware aberration correctors to high-resolution transmission electron microscopy (HRTEM) in the 1990s caused a revolutionary push of this technique and has produced a huge stream of exciting scientific results in this field [1–5]. While the exploration of the new instrumental capabilities for materials science stood clearly in the foreground in the recent years, the practically important aspect of the optical stability in HRTEM gained comparatively little attention [6–8]. This aspect concerns all high-resolution electron microscopes, whether aberration corrected or not, and is in first instance caused by the fact that the allowed deviation of the optical setting from its intended state, which can be still tolerated for a reliable image interpretation, decreases dramatically with increasing resolution. While such optical instabilities hampered already in the past the practical work on the first generation of sub-Ångström microscopes [9,10], their existence can be a strongly limiting factor for reproducible work with the aberration corrected deep-sub-Ångström microscopes of the latest generation, which can access the resolution regime near 0.5 Å [11]. But also for less ambitious work above 1 Å resolution with conventional non-corrected high-resolution microscopes the topic of optical stability cannot be completely ignored.

The reasons for optical instabilities can be subdivided into three principal categories. There exist external influences which can change the optical setting of a microscope. External influences can be user interventions, such as a strong change of lens currents, a change of the object position, or of the object orientation by the goniometer. A different source of external influences can be environmental instabilities, such as temperature fluctuations of the objective lens cooling water, changes of the air temperature, or mechanic floor vibrations. Additionally to these external sources also internal sources, such as electronic instabilities of the voltage and power supplies belonging to optical elements, which are completely independent of user interventions and of environmental influences, can lead to optical instabilities. For the present work the well designed microscope installations of the Ernst Ruska-Centre in Jülich were used with minimal user intervention, so that the observed optical instabilities are almost exclusively due to internal electronic instabilities, whereby minimal environmental components cannot be fully excluded, but are definitely of only minor importance. In summary, the microscopes were investigated in a condition where they were left completely untouched by the user. The investigated scenario is thus close to ideal. Under real working conditions, and on less suited locations, one can thus expect considerably higher instabilities than those reported here.

First indications of significant optical instabilities were already obtained in our previous work, where fluctuations of the twofold astigmatism were monitored with ultra-high precision over long

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[*] Corresponding author at: Peter Grünberg Institute (PGI), Forschungszentrum Jülich GmbH, 52425 Jülich, Germany.

E-mail addresses: ju.barthel@fz-juelich.de (J. Barthel), a.thust@fz-juelich.de (A. Thust).

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timespans using the ATLAS software [12,13]. Following the argumentation of Ref. [12], the twofold astigmatism a_{22} can be seen as a representative marker aberration for the complete optical state of a microscope according to the following reasons: first, the twofold astigmatism and the defocus have as lower-order aberrations the largest influence on the image contrast. Second, whereas the defocus is strongly influenced by the instabilities of the sample holder along the vertical direction, the twofold astigmatism is free of such mechanical influences. Third, the twofold astigmatism as lower-order aberration is in our experience besides the defocus the most volatile aberration, whereas other aberrations tend to be significantly more stable with increasing aberration order. Fourth, the twofold astigmatism can be determined directly in quick sequence allowing a real-time monitoring operation, whereas the determination of higher-order aberrations needs an indirect analysis, which is anyway based again on lower-order measurements.

Since the twofold astigmatism a_{22} is only one of many aberrations adding up to the complete aberration state, the lifetime τ of the complete aberration state is in general shorter than the lifetime τ_{22} of the astigmatism state alone, i.e. $\tau \leq \tau_{22}$. The lifetime τ_{22} of the twofold astigmatism as a marker aberration is thus an optimistic upper limit for the lifetime τ of the complete optical state.

We present in this work a systematic investigation of the optical stability of a variety of different high-resolution electron microscopes operated with different accelerating voltages. Besides the acquisition of large and highly precise astigmatism data from different microscopes, we develop here the theoretical framework for an in-depth statistical evaluation of the acquired data with respect to practically relevant questions, such as the expected lifetime of the optical state on a given microscope under certain working conditions.

2. Experiment

The twofold astigmatism is denoted in the following for simplicity by $a_{22}=a$, the spatial frequency by g, and the electron wavelength by λ. A corresponding lifetime $\tau_{22}(\varphi, g)$ can be defined for any just tolerable deviation φ of the aberration function from its initial value at a targeted spatial frequency g. When applying the widely accepted $\pi/4$ criterion, where one regards a deviation of the aberration function by $\varphi = \pi/4$ as tolerable limit, the corresponding lifetime $\tau_{22}(\pi/4, g)$ is reached for a target frequency g when the twofold astigmatism deviates by an amount of $a_{\pi/4}(g) = 1/(4\lambda g^2)$ from its initial setting [12]. Due to the fact that the $\pi/4$ criterion is at present the sole popular criterion used in electron microscopy, we use without restriction of generality throughout this text a more convenient one-parameter notation, with $\tau_{22}(g) = \tau_{22}(\pi/4, g)$.

In most cases one is interested in the lifetime with respect to the information limit $g_{\max}$, which is the highest linearly transferred spatial frequency [14]. The related lifetime $\tau_{22}(g_{\max})$ can thus serve as a fundamental benchmark figure assessing the optical stability of a microscope. Due to the obvious interest in the optical stability at the information limit $g_{\max}$, the corresponding benchmark figure $\tau_{22}(g_{\max})$ will be addressed in the following also just as the lifetime τ_{22}.

In order to be able to compare microscopes with different information limits $g_{\max}$ one must evaluate the lifetime at one and the same common spatial frequency g, and it can be therefore necessary to evaluate the benchmark parameter $\tau_{22}(g)$ also for frequencies $g \neq g_{\max}$. The case $g < g_{\max}$ concerns practical situations, where one is not interested in exploiting the full resolution offered by the microscope, whereas the case $g > g_{\max}$ addresses the purely theoretical question of how good the stability of a microscope might be in the case of a hypothetical resolution extension beyond the actual information limit.

Table 1
List of investigated microscopes with the applied accelerating voltages U_{acc}, the respective information limit values $g_{\max}$, and the $\pi/4$ tolerances $a_{\pi/4}(g)$ of the twofold astigmatism for $g = g_{\max}$, and for $g = 10\ \mathrm{nm}^{-1}$.

Microscope	U_{acc} [kV]	$g_{\max}$ [nm^{-1}]	$a_{\pi/4}(g_{\max})$ [nm]	$a_{\pi/4}(10\ \mathrm{nm}^{-1})$ [nm]
PICO	50	9	0.58	0.47
	80	12.5	0.38	0.60
	200	18.2	0.30	1.00
	300	15.4	0.54	1.27
Titan	300	12.5	0.81	1.27
Tecnai	200	7.1	1.95	1.00

Long series of coherent high-resolution images of thin amorphous objects were recorded using the following microscopes of the Ernst Ruska-Centre in Jülich:

1. PICO: FEI Titan^3 60–300, C_S and C_C corrected,
2. Titan: FEI Titan 80–300, C_S corrected,
3. Tecnai: FEI Tecnai G^2 F20, uncorrected.

The microscopes were operated with partly different accelerating voltages U_{acc} in a range between 50 and 300 kV, resulting in different values of the information limit $g_{\max}$ for each combination of microscope and accelerating voltage. Table 1 gives an overview over the various experimental scenarios and the according information limit values. Table 1 displays also the tolerances $a_{\pi/4}(g_{\max})$ of the twofold astigmatism, which are allowed by the $\pi/4$ criterion for the respective individual information limit values $g_{\max}$, as well as the tolerances $a_{\pi/4}(10\ \mathrm{nm}^{-1})$ allowed for a common comparison resolution of 1 Å. The latter resolution value of 1 Å is applied regardless whether it can be actually achieved or not.

Image series of amorphous objects were recorded under strict avoidance of any operator intervention on an area of 2048 × 2048 pixels of the installed CCD (charge coupled device) cameras with a binning factor of 4. The microscope magnification and the image defocus were adjusted in such a way that the diffractograms, i.e. the power spectra of the recorded images, exhibited a prominent Thon-ring pattern of at least three rings, which covered the major part of the 512 × 512 pixel diffractogram area [15]. The exposure time of single images was adjusted in the range between 0.25 and 0.5 s. Depending on the read-out speed of the cameras in use it was possible to achieve image acquisition frequencies between 0.5 and 2 Hz. The total series acquisition time, which corresponds to the time window of the stability observation, was chosen to lie in the range between 10 and 75 min. The thereby resulting roughly 1000–4000 images per series were recorded in a fully automated way with the help of an acquisition script. Using the ATLAS software in an automated way in parallel to the image acquisition, the twofold astigmatism belonging to each single image was extracted directly after the image acquisition. By the parallel execution of image acquisition and image evaluation all primary measurement results were available directly at the end of the observation time.

Before presenting the outcome of a typical time series, it is important to state that the directly measured astigmatism is not identical to the true astigmatism, which has been discussed so far in the context of the $\pi/4$ criterion. The difference between the directly measured astigmatism, which is denoted here by $A=(A^x, A^y)$, and the true astigmatism, which is denoted here by $a=(a^x, a^y)$, is due to the error $\delta = (\delta^x, \delta^y)$ of a single astigmatism measurement, which is assumed to follow a normal distribution with respective standard deviations σ^x and σ^y. When inspecting the allowed tolerances of the true astigmatism $a=(a^x, a^y)$ in the third column of Table 1, one finds that these tolerances are in the order of only