Optimized C_S-corrected Imaging of Radiation-sensitive High-resolution Objects

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Often it is radiation damage of the specimen rather than the ultimate resolution of a microscope that determines whether an object can be imaged at a desired resolution. For that reason, individual organic molecules cannot be currently imaged in a TEM. The radiation sensitivity is determined by the sample, current density and microscope acceleration voltage [1] and cannot be significantly improved but it is possible to adjust instrument parameters such that the information per unit electron dose is maximized. In the example below, we assume a "molecule" suspended between tips of an in-situ STM holder, as illustrated in Fig. 1(a). The hydrogen atoms were substituted by iodine, which decreases radiation sensitivity of the molecule and increases image contrast at the iodine sites.

Whether an atom will be detected depends on whether the minimum dose n_0 in $e^-/Å^2$ needed to obtain signal to noise ratio $k=5$ (Rose criterion) is less than the dose which destroys the sample. For bright field (BF) imaging we can write [2] $n_0 > k^2/[f \pi R^2 C^2(R)]$. Here f is the fraction of electrons contributing to background ($f \sim 1$ for BF), $C(R)$ is the contrast and R is the radius of detection area. It is the *average* intensity $I_{avg}(R)$ over a detection area (pixel) with radius R which is detected: $I_{avg}(R) = \frac{2}{R^2} \int_0^R I(r) r \, dr$. The image intensity $I(r)$ can be calculated in the weak phase object approximation with reasonable accuracy at 300 kV [2]. The disadvantage of BF imaging is that low spatial frequencies, where the scattering factor $f(q)$ of an atom is large, are not transferred to image intensity due to the $\sin[\chi(q)]$ term in the contrast transfer function (CTF). The importance of low spatial frequencies can be understood in terms of Fourier-series representation of an object: low-order terms give a strong contribution to signal intensity whereas high-spatial-order terms refine the shape representation but carry less contribution to intensity. The use of Zernike phase plates (ZP) results in transfer of low spatial frequencies to image intensity [3]. In a C_S-corrected TEM equipped with ZP capabilities, the information on the sample may be transferred up to the information limit set by chromatic aberration C_c. Unfortunately, practical implementation of ZP with accurate phase shift and a small enough aperture is not yet viable. An alternative is to measure sample-induced phase shift directly by electron holography, rather than rely on phase transfer to image intensity.

Table 1 and Fig. 1(b) compare the imaging doses n_0 for i) a C_S-corrected TEM in BF, ii) with a $3\pi/2$ ZP and iii) with electron holography. The dose for holography was estimated as $n_0 = 2/[\mu^2 \pi R^2 \phi^2(R)]$, where μ is fringe contrast and $\phi(R)$ is the phase shift as transfered by the microscope and averaged over the detection area of radius R. The choice of 2 in the numerator results in an estimate of the lowest possible limit of n_0 [4]. The fringe contrast was set to 0.75, a rather optimistic value which, however, might be within reach of multiple-biprism set-up in an instrument equipped with a cold field emission gun [5]. Finally we point out that the detection area R has to be kept adequately small ($R \sim 0.3$ Å or less) to obtain sufficiently detailed information to visualize the molecule. This leads to only moderate increase in n_0 compared to the global minimum. We note that optimizing R has only an indirect relation to instrument resolution; it is analogous to optimizing of camera pixel size in

high resolution TEM. Figure 1(b) shows n_0 for H, C, F, Cl, Br, I, Au and U atoms. Considering the accepted radiation damage threshold for aromatic molecules ($n_0 < 100$ e⁻/Å²), the plot indicates that it is not possible to detect light atoms but it may be possible to detect single iodine-substituted organic molecules. It appears that electron holography is an attractive alternative for imaging radiation-sensitive materials [6].

References

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[6] This work was supported by NRC and NSERC in Canada and by the U.S. DOE, BES contract No. DE-AC02-98CH10886.

Table 1. Instrument parameters for R=0.3 Å and n_0 for iodine atom.

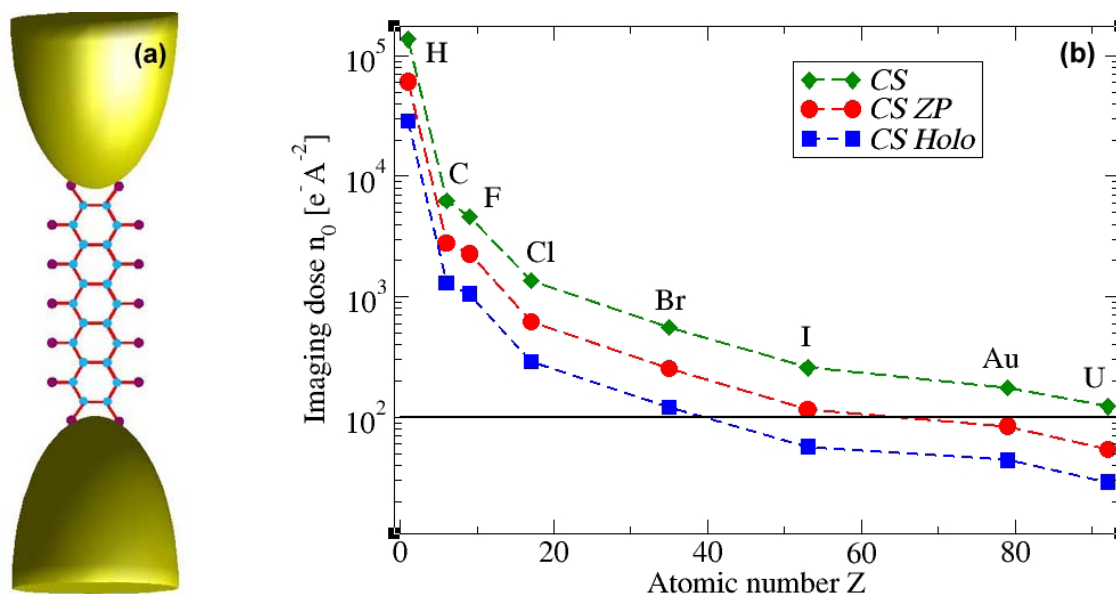| Instrument	U_{acc} [kV]	ΔE_0 [eV]	C_s [μm]	Δz [nm]	C_c [mm]	OL stability [ppm]	HT ripple [eV]	Detection radius R [Å]	Objetive aperture [mrad]	Imaging dose n_0 [e⁻/Å²]	e⁻/pixel @ R=0.3 Å
CS BF	300	0.25	-31	9.0	1.0	0.5	0.1	0.3	24	258	73
CS ZP ($3\pi/2$)	300	0.25	0	0	1.0	0.5	0.1	0.3	>50	98	28
CS Holography	300	0.25	0	0	1.0	0.5	0.1	0.3	>50	57	16

Fig. 1. (a) Schematic experimental set up: an aromatic molecule suspended between tips of an in-situ STM holder; (b) Dose n_0 as function of atomic number Z. Green diamonds: BF in CS TEM; red circles: CS TEM with $3\pi/2$ ZP; blue squares: CS TEM equipped for electron holography. Solid line at 100 e⁻/Å² sets the dose threshold below which an aromatic organic molecule can survive.