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C_s corrected Bright Field TEM Imaging of Radiation Sensitive Materials

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We examined the suitability of C_s-corrected monochromated (CSM) TEMs for bright field (BF) imaging of radiation sensitive materials. In many samples the resolution is set by radiation damage rather than the instrumentation. For example, single organic molecules can not be currently imaged in TEM. In this study we assume a "molecule" suspended between tips of scanning tunneling microscope placed inside a TEM, an experimental set up which may reduce radiation damage by allowing escape of some of the secondary electrons to vacuum rather than damaging sample [1,2]. Ignoring charge redistribution due to bonding, the imaging of single molecules suspended in vacuum is equivalent to imaging ensembles of single atoms. It is known that halogen (such as Iodine) can be substituted for hydrogen in many organic molecules, giving a decrease in radiation sensitivity. An atom can be detected if the dose n_0 in $e^-/Å^2$ needed to obtain signal to noise ratio $k = 5$ (the Rose criterion) is less than the destruction dose. For bright field (BF) imaging $n_0 > k^2/[f\,C^2(R)\;R^2]$ applies [3]. Here f is fraction of electrons contributing to background ($f \sim 1$ for BF), $C(R)$ is the image contrast and R is the detection area radius. For finite pixel sizes, it is the *average* intensity $I_{avg}(R)$ over a detection area (pixel) with radius R which is recorded:

$$I_{avg}(R)=\frac{2}{R^2}\int_0^R I(r)\,r\,dr \quad \text{,where} \qquad I(r)=1+4\pi\int_0^{q_{ap}} f(q)J_0(2\pi r q)q\sin\chi(q)e^{-TD(q)}e^{-SD(q)}dq$$

In this equation, $f(q)$ is the relativistically-corrected atomic scattering factor, $J_0(2\;qr)$ is a Bessel function, (q) is the contrast transfer function (CTF), $TD(q)$ and $SD(q)$ are the temporal and spatial damping functions and q_{ap} is the cutoff spatial frequency corresponding to the objective aperture [3,4]. The CTF was selected for the best imaging conditions of each instrument (negative C_s and positive defocus for corrected and Scherzer conditions for uncorrected instruments [3]). The contrast is defined as $C(R)=|1-I_{avg}(R)|$. Since the dose decreases with illumination angle slowly we set it at 0.1 mrad in our calculations (a reasonable value for field emission microscopes).

Figure 1 shows dependence of n_0 on objective aperture (OA) semiangle $_{ap}$. The oscillations for instrument A originate from oscillations of the CTF; the global minimum is achieved for $_{ap} =$ $_{\text{Scherzer}}$. Instrument B shows a local minimum at $_{ap} =$ $_{\text{Scherzer}}$ but n_0 further decreases when OA is opened more, due to a limited number of oscillations in the CTF. For C, temporal damping removes CTF oscillations and n_0 becomes flat after OA is opened to about $_{ap} = 0.85$ $_{\text{Scherzer}}$. The image intensity profiles (Fig. 2) show more localized contrast for B and C than for A. The dip in the intensity profile for B corresponds to a large OA and a low n_0. The relation between n_0 and detection radius R for I atom (Fig. 3) indicates that CSM instrument may reduce radiation damage from about 330 $e^-/Å^2$ to about 190 $e^-/Å^2$ in BF-TEM of iodine substituted single molecules. This is due to more efficient transfer of the atom phase shift to image intensity for CSM. Figure 4 shows n_0 for H, C, F, Cl, Br, I, At, Au and U. This indicates that for accepted doses for radiation damage of molecules n_0 about 100 $e^-/Å^2$ it is not possible to detect light atoms, but it may be possible to detect iodine-substituted single organic molecules. Techniques that directly measure electron phase (such as electron holography)

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might provide a further decrease in n_0.

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TABLE 1. Instrument parameters

Instrument	U_{acc} [kV]	E_0 [eV]	C_s [mm]	z [nm]	C_c [mm]	*OL stability* [ppm]	*HT ripple* [eV]	*Information Limit* [Å]	*Scherzer angle* [mrad]
A	300	0.5	1	-54	1.5	0.5	0.1	0.75	10
B	300	0.25	-0.0311	9.0	1.5	0.5	0.1	0.62	24
C	100	0.25	-0.0047	4.8	1.5	0.5	0.1	1.17	45

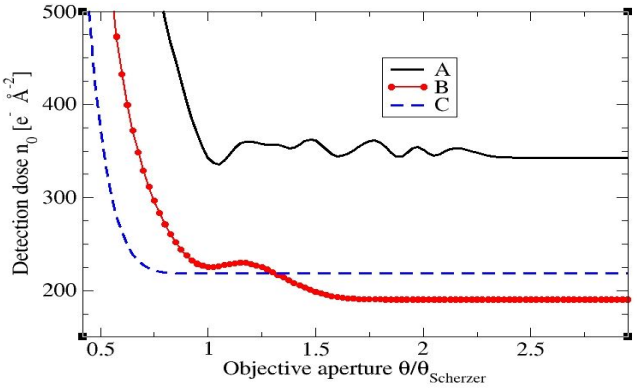

Fig. 1. Detection dose n_0 as a function of the objective aperture.

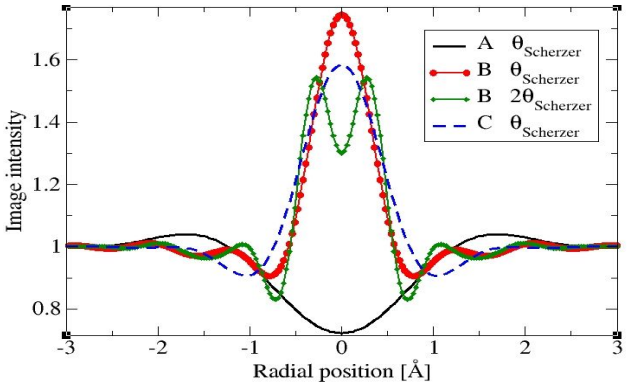

Fig. 2. Image intensity profiles for iodine atom. The illumination angle is 0.1 mrad.

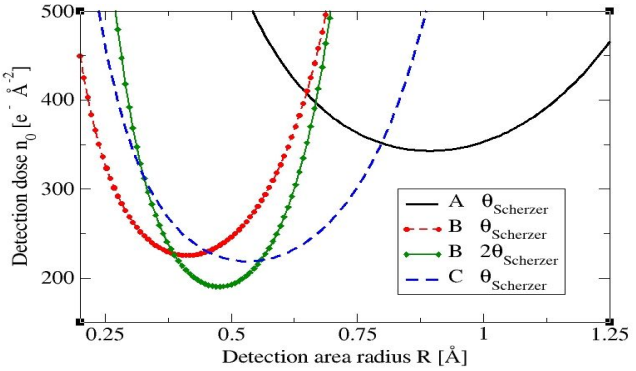

Fig. 3. Dose n_0 needed to detect iodine atom with a detection area with radius R.

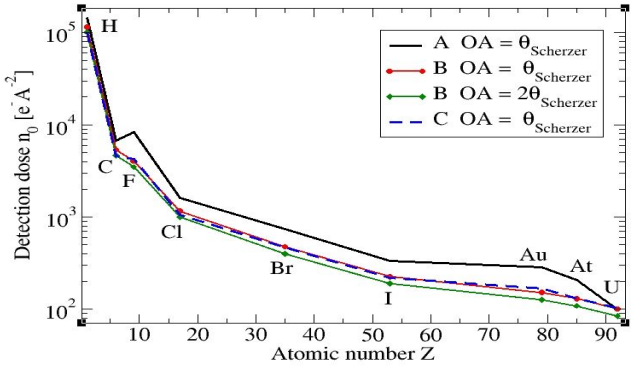

Fig. 4. Dose n_0 as a function of atomic number Z for the minimum n_0 shown in Fig. 3.

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