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Limits to the spatial, energy and momentum resolution of electron energy-loss spectroscopy

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Abstract

We discuss various factors that determine the performance of electron energy-loss spectroscopy (EELS) and energy-filtered (EFTEM) imaging in a transmission electron microscope. Some of these factors are instrumental and have undergone substantial improvement in recent years, including the development of electron monochromators and aberration correctors. Others, such as radiation damage, delocalization of inelastic scattering and beam broadening in the specimen, derive from basic physics and are likely to remain as limitations. To aid the experimentalist, analytical expressions are given for beam broadening, delocalization length, energy broadening due to core-hole and excited-electron lifetimes, and for the momentum resolution in angle-resolved EELS.
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1. Introduction

Many considerations affect the capability of electron energy-loss spectroscopy (EELS), in combination with transmission electron microscopy (TEM), to solve problems in materials or life science. Some of them relate to instrument design, such as the electron-optical design of the spectrometer and microscope column. Others are partly environmental, such as the electrical and mechanical stability. There are also important human considerations, including the knowledge, skill and patience of the researcher. But over the last few decades, the knowledge base and level of instrument performance have improved to the extent that a third kind of factor becomes important: performance limits arising from basic physical principles. This paper reviews all of the relevant factors but gives emphasis to these fundamental limitations. Although the physical principles involved are well established, they are presented here in a form designed to be convenient for the experimentalist.

2. Spatial resolution

In EELS, spatial resolution refers to the smallest diameter (lateral dimension within a thin specimen) from which spatial information can be obtained. In energy-filtered (EFTEM) imaging, the equivalent quantity is the minimum useful pixel size, below which there is no substantial gain in information content.

2.1. Electron-optical considerations

Because spectroscopy and scanning-transmission (STEM) imaging are usually carried out using a tightly focused beam (electron probe), one obvious limit is the smallest beam size that can be produced by a given instrument. In a modern TEM, the minimum probe size is well below 1 nm, thanks to the efficient exploitation of electromagnetic lenses. Use of a Schottky or a cold field-

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emission (CFE) electron source helps to ensure that there is sufficient current (e.g. 1 nA) in such a small probe. Correction of the spherical aberration of the probe-forming lens allows a further increase in probe current and can reduce also the full width at half maximum (FWHM) of the probe to below 0.1 nm [1–3]. By removing aberration tails that otherwise contain a substantial proportion of the current, the corrector should provide a "cleaner" current-density profile.

Although the probe size is determined by the electron-source size, lens aberrations and electron diffraction at the condenser aperture, it can in principle be degraded by Coulomb interactions between the electrons within the TEM illumination system. Fortunately, the TEM uses a relatively low beam current, giving conditions that correspond to the "pencil-beam regime" where the statistical Coulomb broadening depends on the third power of the beam current [4]. So for accelerated electrons, the statistical broadening appears to be negligible, even for aberration-corrected lenses forming a high-intensity probe, where the current density can exceed $1\,\mathrm{MA/cm^2}$.

2.2. *Beam broadening within the specimen*

Inside the specimen, the electron beam spreads laterally due to electron scattering. Even without scattering, it would broaden by $2\alpha t$ (of the order of 1 nm for a tightly focused probe and thin specimen) for a probe of convergence semi-angle α focused onto the top surface of a specimen of thickness t. For an amorphous specimen, elastic scattering increases the beam broadening by an amount [5]:

$$b \approx (625\,\mathrm{cm})(Z/E_0)(\rho/A)^{1/2}[t(\mathrm{cm})]^{3/2}$$
$$\approx (0.47\,\mathrm{nm})(\rho Z)^{1/2}(100\,\mathrm{keV}/E_0)[t/50\,\mathrm{nm}]^{3/2}, \quad (1)$$

where ρ is the specimen density in $\mathrm{g/cm^3}$, E_0 the incident-electron energy in keV. For $E_0 = 100\,\mathrm{keV}$ and $t = 50\,\mathrm{nm}$, $b = 1.8\,\mathrm{nm}$ for carbon, 2.9 nm for Al, 7.6 nm for Cu and 17 nm for Au, values that agree rather well with the diameter (containing 90% of the trajectories) deduced from Monte Carlo calculations [5]. This effect may be added in quadrature to the beam-divergence effect ($2at$) in a first approximation.

Although beam broadening in the specimen largely determines the spatial resolution of X-ray energy-dispersive spectroscopy (XEDS), its effect on transmission EELS can be considerably less, *provided* the spectrum is recorded through an angle-limiting aperture that removes electrons that deviate from the optic axis by more than its semiangle β. Considering scattering at a distance z above the bottom surface of the specimen, electrons entering the spectrometer travel through a cone of radius βz and volume $(\pi/3)\beta^2 z^3$. Averaging over the specimen thickness t, the diameter containing n% of the detected electrons is:

$$d_n \approx F_n \beta t, \quad (2)$$

where $F_n \approx 0.4$ for $n = 50$ and $F_n \approx 1.0$ for $n = 90$ [6]. Taking $t = 50\,\mathrm{nm}$ and $\beta = 10\,\mathrm{mrad}$ gives $d_{50} \approx 0.2\,\mathrm{nm}$ and $d_{90} \approx 0.5\,\mathrm{nm}$, values considerably less than the total broadening given by Eq. (1). These estimates may actually be pessimistic since they assume that the scattering (per unit solid angle) is constant up to the angle β. If no angle-limiting aperture is used, the beam width in EELS should be given in Eq. (1), at least for an amorphous specimen.

For crystalline specimens, a more correct treatment of beam broadening includes the fact that elastic scattering (except in ultra-thin specimens) is dynamical: for depths in excess of $\xi_g/2$, where ξ_g is the extinction distance (in the range 25–100 nm for 100 keV electrons), many electrons are scattered back *towards* the optic axis. As a result, the electron beam spreads less than in an amorphous material (at least for scattering through less than a typical Bragg angle), which benefits XEDS analysis in addition to EELS [7,8]. In addition, the electron density within the beam becomes distributed non-uniformly: at depths exceeding about 5 nm, electrons are channeled preferentially along the columns of atoms, especially for a crystal oriented with a low-index zone axis parallel to the incident beam [9].

2.3. *Chromatic aberration*

If the electrons transmitted through a specimen are subsequently focused to form an energy-filtered (EFTEM) image, the image resolution is subject to degradation by lens aberrations. Usually chromatic aberration is the most important factor, particularly for core-loss images where the energy and angular widths of the focused electrons can be considerable. Assuming the energy-filtered image is correctly focused for electrons that pass though the center of the energy-selecting slit, the diameter containing 50% of the electrons is increased by an amount Fd_c, where $d_c = C_c\beta(\Delta/E_0)$, C_c is the chromatic aberration coefficient of the objective lens and Δ is the energy width of the energy-selecting slit. The factor F depends on the angular width of the inelastic scattering: $F \approx 0.1$ for an energy loss $E = 100\,\mathrm{eV}$ and $E_0 = 100\,\mathrm{keV}$, increasing to 0.3 for large E or a thick specimen [10]. The diameter Fd_c is typically around 0.2 nm for $\beta = 10\,\mathrm{mrad}$ and $\Delta = 20\,\mathrm{eV}$. Note that this 50% broadening is substantially less (by factor F) than the *total* chromatic width d_c, which is often taken as an estimate of the chromatic effect.

In principle, the EFTEM resolution also depends on the spatial resolution of the electron detector (usually a scintillator/CCD arrangement). However, this factor can be made unimportant by choosing a sufficiently high image magnification.

None of the above factors are fundamental, in the sense that they depend on the design of the microscope and

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