Towards sub-10 meV energy resolution STEM-EELS

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Abstract. A monochromator we have introduced is improving the attainable energy resolution of electron energy loss spectroscopy (EELS) in a scanning transmission electron microscope (STEM) by more than 2x relative to what has been available until recently. Here we briefly review the design and the performance attained so far. We then investigate the ultimate resolution limits of our system and show that it should be able to reach an energy resolution of <10 meV.

1. Introduction
The high energy resolution monochromated EELS-STEM (HERMES™) system we have recently developed has introduced several new design concepts [1-3]. It has been able to reach 12 meV energy resolution (full-width at half maximum (FWHM) of the zero loss peak (ZLP)) in a short-exposure spectrum [2] and <20 meV in longer-exposure ones (e.g. Fig. 1). It can analyze samples at atomic or near-atomic resolution. When monochromating to about 100 meV, it has slightly improved the spatial resolution of the STEM it has been built into [2].

Fig. 1. Zero loss EELS peak obtained with Nion HERMES™, using a Gatan Enfinium EELS [4] equipped with ultra-high stability multipole power supplies. 100 keV, 1 s total acquisition.

In this paper, we review the reasons for the excellent energy resolution reached already, and we discuss the ultimate limits of the system.

2. Brief description of HERMES™
The system’s monochromator uses an α-type electron trajectory, in which the beam traverses three magnetic prisms, the first one being traversed twice. The monochromator is located in the main column of the microscope, outside the electron gun. It energy-disperses the electron beam.
incident on a slit located in the mid-plane of the monochromator, and then undisperses it such that the beam re-inserted into the column has no energy dispersion in position or in angle.

Quadrupoles in the pre-slit half of the monochromator magnify the dispersion of the energy spectrum produced by the first prism, by an adjustable amount up to several hundred µm/eV (at 100 keV), and project the spectrum onto the energy-selecting slit. A post-slit set of quadrupoles, arranged symmetrically to the pre-slit set, works together with the system’s prisms to undo the dispersion as the beam is steered back into the column. The electron trajectories fulfil precise symmetry requirements at the energy-selecting slit [1], without which complete un-dispersion would not be possible. Sextupoles and octupoles are provided so that important second and third order aberrations can be cancelled both at the slit and in the beam re-entering the microscope column.

3. Improved energy stability

The principal reason for the improved energy resolution of our system is that unlike most other designs, our monochromator produces an electron beam whose energy does not change when the high tension (HT) of the microscope changes. HT is difficult to stabilize to better than about 0.3 parts per million, i.e. about 30 mV r.m.s. for a 100 kV power supply. If the monochromator is located in the gun, upstream of the electron accelerator, and the spectrometer sits at ground potential, as is done in most monochromator designs, changes in the HT shift the EEL spectrum. In our design, both the monochromator and the spectrometer sit at ground potential. A change in the HT merely shifts the dispersed beam on the monochromator’s energy-selecting slit, and the energy of the beam admitted into the microscope column and the position of the spectrum on the EELS CCD do not change.

Fig. 2. Schematic diagram illustrating how various instabilities affect the HERMES™ energy resolution. MC = monochromator, P1 to P4 = prisms of the system. The microscope column is represented only by the condenser-objective lens, with a thin sample in its middle. Downward-pointing arrows mark the energy admitted by the energy-selecting slit.

Fig. 2(a) illustrates the normal operation of the monochromator: the energy-dispersed beam is centred on the slit, and only the central range of electron energies (green online) is admitted into the microscope column. The three magnetic prisms of the monochromator (P1 to P3) are connected in series. The prism of the electron energy loss spectrometer (P4) is also connected...
in series with the other prisms, and the “green” electrons arrive at the normal ZLP position on
the EELS CCD. Electrons that have lost energy in the sample arrive on the CCD at a different
position, forming a dispersed spectrum.

If the HT value changes, say by +200 mV, while the prism current remains the same in our
system (Fig. 2(b)), the energy-dispersed beam at the slit shifts in position by an amount
Corresponding to an “energy gain” of 200 meV, as shown in the figure. The slit keeps on
selecting the same energy, and the rest of the microscope column operates on an electron beam
of exactly the same energy as before. Because the energy distribution of the cold field emission
gun used by our system is rather narrow, changes in HT of as little as 20 mV can result in an
intensity change of the beam admitted through the slit. We minimize such changes by sensing
the current incident on the two slit halves, and using the difference signal in a feedback scheme
controlling the fast loop of the HT generator so that the beam remains centred on the slit.

Fig 2(c) and (d) illustrate what happens when the monochromator prism current changes.
Low-power current supplies are straightforward to stabilize to about 0.05 ppm r.m.s. (or better),
but even such a small change alters the energy of the selected electrons by about 10 meV at 100
keV. A change in the energy of the selected beam due to a change of the prism current is
indicated in the figure by the beam becoming red (online). If the monochromator prisms and
the EELS prism were powered by separate power supplies (Fig. 2(c)), the EELS spectrum
would shift on the detector and the energy resolution in a time-integrated spectrum would
become worse. A change in a separate current in the EELS prism would have a similar effect.
However, when the EELS prism is supplied by the current that runs through the monochromator
prisms (Fig. 2(d)), then the magnetic field in the EELS prism also changes, and the position of
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In reality, of course, the four prisms of the system cannot be tied together rigidly if the
ability to change the prism excitations independently by small amounts is to be retained. Prisms
2-4 are therefore provided with weak auxiliary windings, whose power supplies run outside of
the stabilization scheme. The magnetic fields produced by these windings are only about 3% of
the main fields, and changes in their contributions to the total fields of the prisms thus produce
beam deflections that are some 30x smaller than deflections due to fully independent prisms.

In our design, only instabilities that enter the microscope column between the slit of the
monochromator and the EELS detector can directly affect the EELS energy resolution.
Instabilities that enter the column between the MC slit and the sample (i.e., instabilities in the
exit half of the MC, condenser lenses and their deflectors, corrector, quadrupole lens module,
scan coils and the objective lens) also shift the probe on the sample and thus cause a loss of
spatial resolution. Because of this sensitivity, the pre-sample part of the column is protected by
several stability-enhancing measures, such as a mechanically rigid construction, triple and
quadruple magnetic shielding, and highly stabilized power supplies. The post-sample part of
the column, on the other hand, offers no similarly independent stability diagnostic, and EELS
spectrometers up to now have typically only dealt with incident beams 50 meV and more in
energy width. As a result, the rigorous stability measures of the pre-sample column are not
always followed: the magnetic shielding tends to be single-layer, the detector column and the
EELS are typically less rigid mechanically than the main column, and the power supplies are
not always designed for the few-parts-per-10^8 stability that pre-sample power supplies aim for.

Fig. 3 illustrates the improvement possible when the performance of an important part of the
post-column optics is improved in a stabilized monochromator system. It shows the position of
the zero loss peak on the CCD detector of the Gatan Enfinium EELS, measured at 100 keV for
a series of spectra recorded at about 100 spectra a second, and plotted as a function of time.
Trace (a) shows the ZLP positions when the Enfinium multipoles are powered by Gatan’s
regular current supplies, trace (b) shows the positions when Gatan’s newly developed “ultra-
high stability” supplies are used.
The peak-to-peak variation is about 50 meV in trace (a) and about 12 meV in trace (b). The four-fold improvement in the stability meant that spectra acquired in 0.1 s with the improved power supplies gave ZLPs 17 meV wide (and 18 meV wide for 1 s acquisition), whereas spectra acquired in 0.1 s with the regular power supplies gave ZLPs 34 meV wide. The larger broadening would clearly not be of concern if the HT of the microscope caused say a 50 meV instability, or if no monochromator was being used. In the present case, however, the regular EELS power supplies were a major limit on the performance of the total system, and even the improved power supplies may not be sufficiently stable if the energy resolution is to be improved to <10 meV.

![Graph showing variation in the position of the zero loss peak on the EELS CCD as a function of time.](image)

**Fig. 3.** Variation in the position of the zero loss peak on the EELS CCD as a function of time. a) regular Gatan Enfinium multipole power supplies, b) ultra-stable power supplies.

### 4. Fundamental limits on HERMES™ performance

The energy resolution of the monochromator and of the electron spectrometer is ultimately determined by the ratio of their energy dispersion to the size of the monochromatic beam crossover (measured at the monochromator slit and at the EELS detector, respectively).

As shown schematically in Fig. 2, the electron beam in our monochromated STEM-EELS system goes through a number of crossovers. The crossovers are simply successive images of the cold field emission source: highly magnified and energy-dispersed at the monochromator’s slit and the EELS detector, highly demagnified and undispersed at the sample. Three of the crossovers play a fundamental role in determining the performance of the system. The crossover at the sample determines the spatial resolution, and the crossovers at the monochromator slit and at the EELS detector together determine the energy resolution. For each one of these crossovers, the range of the incident angles is set by an aperture whose size is chosen such that the crossover is not broadened by aberrations. The crossover size is then determined by diffraction due to the chosen aperture and by the chosen beam current, which is proportional to the projected area (magnification<sup>2</sup>) of the source.

Many other factors can increase the crossover size: magnetic, electric and mechanical instabilities, charging of apertures or of the energy-selecting slit, stochastic Coulomb repulsion, and magnetic Johnson-Nyquist noise arising due to thermal motion of electrons in the drift tube, which has been pointed out recently (5,6). Provided that these effects are suitably minimized, the size of each crossover is given by:

\[
d = 0.61 \left( \frac{\lambda}{\alpha} \right) \left( 1 + \frac{I_b}{I_c} \right)^{0.5}
\]

where \(\lambda\) is the electron wavelength, \(\alpha\) the semi-angle of the electron beam converging on the crossover, \(I_b\) the beam current at the crossover and \(I_c\) the coherent current of the electron source.

The electron-optical limit on the attainable energy resolution of the monochromator and the spectrometer is given by

\[
\delta > \frac{d}{\zeta}
\]
where $\delta$ is the energy resolution and $\zeta$ the energy dispersion of the spectrum in each device. The energy resolution is of course also broadened by the finite width of the slit in the monochromator and the finite spatial resolution of the EELS detector. However, the influence of these factors can be lessened by increasing the dispersion at the slit and at the detector, and they do not need to be discussed when evaluating the fundamental limits.

$\delta$ is determined most simply by examining the spectrum produced by the principal energy-dispersing element: prism 1 of the monochromator and prism 4 of the spectrometer. This is because subsequent magnification of the first spectrum by the system’s quadrupoles magnifies the crossover and the energy dispersion by the same amount, and therefore does not change the energy resolution. The energy dispersions of the MC (Nion) and the EELS (Gatan Enfinium) prisms are 0.9 $\mu$m/eV for the main monochromator prism, and 0.8 $\mu$m/eV for the EELS one (at 100 kV). The preferred radius of the beam entering the MC in our present set-up is $\sim 100 \, \mu$m, which gives, for a beam current equal to $3 \, I_c$, a crossover 7 nm in size formed in the (virtual) spectrum just after prism 1, and an energy resolution limit of 8 meV. For the EELS, our preferred entrance beam radius is 500 $\mu$m, and the beam current, which is limited to less than the monochromator value by a virtual objective aperture (VOA) positioned after the MC, is roughly equal to $I_c$ (when the MC slit is open wide). This gives a theoretical EELS prism spectrum crossover size of 1.6 nm and an energy resolution limit of 2 meV. In other words, both the systems are capable of giving an energy resolution of 10 meV and better. We have not yet progressed to this level, most likely due to the individual limits all adding up (in quadrature) and due to remaining instabilities.

The EEL spectrometer comes off better in the above comparison, for two simple reasons: we admit a wider diameter beam into it than into the MC, and we restrict the beam current entering it more than the current entering the MC. The large entrance beam places strict requirements on the quality of the aberration correction performed in the spectrometer, but these can be readily met by an aberration-corrected spectrometer, as shown for instance in Fig. 7 of [2]. Fortunately, because the electrons stop at the EELS detector, a poor EELS set-up affects only the EELS energy resolution and no other aspects of the microscope performance. The EELS set-up is also simplified by the fact that only aberrations in the dispersion direction affect the quality of the EEL spectra.

On the minus side for the EELS, the phase space occupied by the electron beam can be greatly increased by scattering at the sample, particularly when the fast electrons are scattered to large angles (as they are when exciting phonons) and the sample is illuminated by a broadly defocused probe (to minimize radiation damage). In this case, the whole area illuminated on the sample becomes “a new source” that is imaged onto the EELS detector, and excellent energy resolution is likely to become impossible.

For the monochromator, aberrations in both the dispersion direction and perpendicular to it need to be controlled, because the crossover imaged at the MC slit is re-imaged onto the sample. Monochromator aberrations being able to spoil the probe-forming performance of the microscope is why we do not allow the beam entering the monochromator to be as wide as the beam entering the EELS. If we are to improve the energy resolution of our whole system to $\sim 5$ meV, we will need to work with an electron beam that is more than 300 $\mu$m wide (in the dispersion direction) in the monochromator’s main prism (at 100 keV). Such a beam will be rather sensitive to stray magnetic fields and instabilities due to charging, but it should nevertheless be manageable with greater experience in monochromator tuning.

The path to energy resolution better than 10 meV in our system is thus rather clear: we have to clean up all remaining instabilities, and we have to improve the quality of the tuning of the monochromator and the spectrometer so that even the wide beams needed to minimize the diffraction limit do not give rise to appreciable aberrations. It is also worth noting that because the dispersion of magnetic prisms is proportional (for non-relativistic electrons) to $1/E_0$, (where $E_0$ is the primary energy) whereas the diffraction-limited size of a crossover only grows as
1/√E₀, the energy resolution of our system is expected to be proportional to √E₀. In other words, the energy resolution at 30 keV should be nearly 2x better than at 100 keV.

Another limit we need to be concerned about comes from the brightness of the electron source, as discussed in [2]. With an energy width of the electron source of around 300 meV (as is typical for W CFEG) and an energy interval of say 10 meV selected by the slit, 97% of the beam electrons are intercepted by the MC slit even when the slit and the focusing of the spectrum on it are absolutely perfect. A beam current of 300 pA entering the monochromator then gives a probe current of <10 pA on the sample. To maintain a bigger current the probe then has to become larger, i.e., we must give up spatial resolution in order to have a more substantial probe current at a very good energy resolution. A brighter / more monoenergetic electron source than tungsten CFEG would decrease this difficulty.

One more limit to which we are paying close attention is the extent of the “skirt” of the zero loss peak (also called “tail”) that can arise due to effects such as mistuned aberrations, aperture charging, and the sideways spreading of the signal in the detector. Our goal is to reduce the skirt to <10⁻⁵ of the ZLP’s maximum intensity just 100 meV away from the maximum, and we are pursuing several measures to achieve this.

5. Conclusion
When the monochromator and the electron spectrometer overcome instabilities well enough to become diffraction-limited, better energy resolution will require that the beam entering either device is made larger. We are now approaching this limit in the Nion monochromator, and we are likely to reach it soon in the energy loss spectrometer too. We will then have entered the realm of “diffraction-limited EELS”, in which the whole STEM-EELS system will be limited by the quality of its electron optics rather than by the energy spread of the electron source or the instabilities of the system. This type of performance limit is rather familiar to us: it is similar to the STEM spatial resolution limit due to spherical aberration, which we were able to improve substantially by developing a STEM aberration corrector [7,8].

A STEM-EELS system operating at this advanced level is likely to be able to attain <10 meV energy resolution with an atom-sized (<2 Å) electron probe, and thereby to open up a new field for experimental study: phonon spectroscopy with atomic spatial resolution. It is not every day that a new type of physical interaction becomes available in the electron microscope. It promises to make our efforts to improve the energy resolution further very worthwhile, every step of the way.

Acknowledgement
We are grateful to Profs R.W. Carpenter and P. Rez for stimulating discussions, and to Dr. C. Trevor of Gatan Inc. for loaning us ultra-high stability power supplies and for providing a DigitalMicrograph™ script that analyzes the ZLP positions in a sequence of EEL spectra. Financial support was provided by NSF grant DMR MRI-R2 #959905, DOE grant DE-SC0007694, and by Rutgers University and Nion Co.

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