An energy stabilizing system for electron energy-loss spectrometers in transmission electron microscopy

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Abstract

An energy stabilizing system for electron energy loss spectrometers in a transmission electron microscope (TEM) is described. The key element is a wire sensor based fast electron detector that can sample the 'zero-loss' component of the transmitted electrons at a high refreshing rate. A digital based real-time processor is used to determine the relative shift in the zero-loss component beam and to produce a closed feedback loop to stabilize the energy dispersed electron distribution at the back focal plane of the post-column spectrometer. The system had the distinctive advantage of simultaneously acquiring a drift corrected electron energy loss spectrum over the energy range where the beam is sensed while stabilizing the whole energy loss distribution for more efficient acquisition by a separate (parallel) detector. A TEM version of the alignment figure is described to aid the alignment and operation of the system for an energy stabilized post-column spectrometer in a conventional transmission electron microscope. The effect of aberration on the accuracy of the energy stabilization is discussed.

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1. Introduction

Electron energy loss spectroscopy has firmly established itself as an extremely useful analytical technique which can have single atom sensitivity [1]. The information content in EELS is proportional to the energy resolution achieved. For example, the EELS resolution in the order of 2 eV or larger is good enough to distinguish between different absorption edges of the atoms present, hence it can be used to determine the chemical composition [2]. The EELS resolution between 0.5-2 eV is routinely achieved with the most of the current generation analytical electron microscopes. This allows different chemical bonding configuration, for example between the sp\textsuperscript{2} and sp\textsuperscript{3} bonding in carbons [3] or the valence state of transition metals [4] to be distinguished, as well as the bandgap of some wide bandgap materials to be determined [5]. Compared with X-ray based spectroscopy, electron beams can be focused

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down to a sub-nanometre-sized probe or atomic resolution images can be formed using electron lenses [6]. This allows chemical and electronic information obtained with atomic spatial resolution. It is expected that further improvement in the achievable electron energy loss spectrosopes will deliver more useful analytical information. It is therefore a challenge to devise electron optical system which can deliver EELS with finer energy resolution.

The practical energy resolution of electron energy-loss spectrometers (EELS) for use in transmission electron microscopes (TEM) is determined by a combination of factors such as the energy spread of the electron source, the aberration of the spectrometer, the instabilities of the electron accelerating voltages as well as mechanical vibrations. Progress has been made with the improvement in the electron source technology from thermionic to field emission source. Now with the introduction of source monochromators, the energy spread can be reduced to less than 10 meV for a transmission electron microscopy operating at 200keV [7]. However, in many practically useful cases, the routine achievement of good energy resolution, particularly over extended recording period, is limited by fluctuation in the high tension of the electron microscope [8]. In this paper, we will describe an energy-stabilized electron energy-loss spectrometer for use in a transmission electron microscope.

2. Design goal and practical layout

The design goal is to detect the drift of the energy distribution of the electron beam at the energy-dispersion plane of the spectrometer. This is achieved by a fast-response electron detector to track the movement (or drift) in the most intense portion of the energy dispersed pattern in the electron beam (usually dominated by electrons that have lost negligible energy, or the so-called zero-loss peak in EELS). The detected drift signal is used in the feedback loop to stabilize the position of the whole energy dispersed electron beam pattern. A separate detector, focusing on a separate portion of the energy dispersed pattern of the electron beam, can take advantage of the energy stabilization, to record the spectral details in its best resolution (i.e. that determined by the energy spread of its electron source). Fig. 1 shows a schematic block diagram of one such implementation we used for a post-column prism spectrometer which is also included in the diagram, although the design principle should be applicable for other types of spectrometers such as the omega type spectrometer [9]. The fast-response detector is a wire-sensor based system and the second detector is a slow scan charge-coupled-device (CCD) camera. A multiplet-based lens system is inserted before the CCD camera for magnification so that the pixel size of the CCD is best matched with the energy dispersion of the electron beam pattern after the spectrometer.

2.1. Wire-sensor based fast response detector

The fast response detector is the crucial element of the energy stabilizing system. It not only needs to sense the drift of the electron beam pattern, but it also should allow undisturbed transmission of the spectral regions of interest so that they can be detected by the other detector. A wire-sensor based fast-response detector can serve this purpose very well [10]. Fig. 2 shows the design and practical implementation of a wire-based electron beam position detector. Its core is a tungsten wire of about five micrometer diameter stung across a metal slot that forms part of the electron beam drift tube. The position of the electron beam is determined by interacting with the wire. The backscattered electrons arising from beam-wire interaction is detected by a scintillator whose light emission is guided by a light tube to a photomultiplier tube (PMT) attached to the exterior of the beam position detector. With the wire detecting the strongest signal in the energy dispersed beam, the upper slot in the wire detector can allow the passage of the electron beam of higher energy loss to pass through to a CCD-based parallel detector. By stabilizing the position of the strongest beam with respect to the wire, we can achieve the energy stabilized detection of the electron energy loss spectrum at the parallel detector.

2.2. A digital real-time feedback and spectrum acquisition system

A unique feature of our drift detection and feedback system is that the operation is digitally based. This gives us a great deal of flexibility, for example, in fine tuning the feedback system as well as simultaneously recording of the spectral information over the energy range where the sensing is taking place. The detailed construction of the digital
feedback system will be reported elsewhere, but the main architecture can be summarized as follows. The energy dispersed electron beam at the spectrum dispersion plane of the prism spectrometer is made to oscillate about the position of the wire sensor. For the benefit of the post-wire-sensor parallel detector, the dispersed beam will be demodulated afterwards by the same oscillating signal. The periodically time-varying signal from the PMT is digitalized and recorded as a serially acquired energy loss spectrum by a National Instrument PXI-based real time system. The spectrum drifts for successful oscillating periods are measured and compared with respect to the spectrum acquired at the first oscillating period. The drift signals are fed to a PID controller and the output is added to the beam oscillating signal. At the same time, successively acquired spectra are also drift corrected digitally and an accumulated spectrum is recorded within the PXI-system and passed on to the host computer for further detailed analysis.

Fig. 1. Schematic block diagram of the energy stabilized post-column electron energy-loss spectrometer for transmission electron microscopy.

3. Alignments and preliminary results

3.1. Alignment

For the correct operation of the wire sensor, the correct alignment of the electron optics in its front is crucial, so the wire detector is intersecting a least aberrated electron beam. In many ways, the formation of the spectrum from a wire-sensor is closely related to the serial acquisition of EELS using a slit-based detector in the place of the wire-detector [11], so we initially have been using the traditional procedure to align the wire sensor, i.e. to fine tune the relevant spectrometer controls such that the narrowest zero-loss peak is produced. In our implementation, we have used a prism spectrometer from the Gatan 607 serial EELS system originally equipped for a Philip 430 transmission electron microscope. It comes with two quadruple lenses and a sextuple lens at the entrance side of the prism.
spectrometer to provide first order focus and a rough second order aberration correction [12]. These auxiliary lenses are adjusted to project the image at the back focal plane of the projector lens to the wire sensor at the spectrometer dispersion plane. Other factors in the TEM can also affect the spectrometer alignment, for example, the diffraction focus control which controls the cross-over position of the back focal plane of the projector lens along the optical axis in the diffraction coupled mode of operation.

Alignment of the spectrometer and the project lens of the transmission electron microscope is a multivariable search process and the result may not be the global minimum if a single criterion such as the width of the zero-loss beam is used. In scanning transmission electron microscope (STEM) without a sophisticated post-specimen lens system, the alignment of the electron optics can be aided by forming a so-called 'alignment figure' which is the mapping of the intensity passing through a narrow energy selecting slit at the spectrometer dispersion plane as a function of the angle of incidence at the spectrometer [11]. In a conventional transmission electron microscope (CTEM) with its complicated post-specimen optics, such an aberration figure can not be easily recreated. Instead, we have added a pair of scanning signals to the deflection coils for the final projector lens and form a ‘TEM alignment figure’ by mapping the intensity detected at the wire sensor as a function of the angle of deflection. Fig. 3 shows the optical ray diagram of the projector lens and that of the prism spectrometer up to the position of the wire detector. A change in the projection lens deflection translates largely as an image shift at the back focal plane of the projector lens. As the back focal plane of the projector lens is the object plane of the spectrometer, our ‘TEM alignment figure’ is equivalent to scanning of the beam crossover at the back focal plane of the projector lens while detecting the intensity of the beam intersected by the wire-sensor. The same result will be obtained if the wire sensor is replaced by a slit sensor in a serially acquired EELS system.

At least two ways can be found for such a figure to be used for determining the electro-optical parameter in our set-up. When there is no specimen in the column of the electron beam, the dispersed electron beam pattern consist the zero loss beams only. If we adjust the current in the spectrometer such that the zero loss beam just intersect the wire sensor, then the ‘alignment figure’ is a figure which can be understood as the convolution of the beam shape with the interior aperture shape at the vacuum valve isolating the beam drift tube from the TEM camera chamber. A series of such aberration figures as a function of the ‘diffraction focus’ (i.e. under different excitation of the intermediate lens) is shown in Fig. 4. When the beam cross-over coincides with the position of the vacuum isolation valve, we have the smallest alignment figure and the highest beam transmittance. This gives us an excellent experimental method of determining the proper intermediate lens excitation. This can also be used to center the beam cross-over with respect to the aperture. Once these are fixed, we can generate a series of alignment figures by changing the current in the main bending magnet. In the prism spectrometer employed, the main purpose of the hexapole coils is used to correct the second order aberration. To show that the ‘TEM alignment figure’ is sensitive
to the second order aberration, we have recorded such ‘alignment’ figures as a function of the excitation of the hexapole coils about the value ($I=0.85\,\text{A}$) responding to the optimal energy resolution of electron spectrum in Fig. 5. This sensitivity to both the first order and second order change in the electron optical parameters opens the way for the alignment figure to be processed as an on-line aberration diagnostic tool for real time automatic tuning.

Fig. 3. The optical ray diagram for the generation of the alignment figure.

Fig. 4. The effect of changing the diffraction focus to the aberration figure in JEM 2000EX. (a), (b), (c), (d), (e) correspond to different sizes of the diffraction focus.

Fig. 5. Alignment figures obtained by scanning the deflection coils above the projector lens (see the text for details). From (a) to (c), the hexapole coils current ($I$) is systematically changed around the value corresponding to the optimal energy resolution of the electron spectrometer.
3.2. The nature of energy instability

Our system, together with the prism spectrometer has been tested out in a JEM-2000EX TEM equipped with a thermionic tungsten electron source. As such, we can not test the energy resolution of the spectrometer to its limit. But by monitoring the drift error signal before the energy stabilization is applied (an example is shown in Fig.6) we can study the temporal fluctuation of the zero-loss peak at the exit focal plane of the post-column electron energy analyzer. In the short period of 30 seconds, the zero-loss peak has already fluctuated over an energy range around 4 eV. However, we are able to track the zero-loss peak shift much smaller than the width of the zero-loss peak (which is about 3.5 eV) for a long period of time (more than 24 hours has been achieved).

3.3. The energy stabilized electron energy loss spectrum

Fig.7 shows a drift corrected EEL spectrum where the oscillating signal is provided by a Tektronix arbitrary function generator connected to the scanning coils of the prism spectrometer (alternatively the drift tube voltage). The electron energy loss spectrum acquired using this method can span over 150 eV. In this case, the electron beam is passing through a thin flake of silicon single crystal prepared by ion beam thinning. The EELS spectrum is dominated by the ‘zero-loss’ beam followed by evenly spaced peaks of diminishing intensity up to 100 eV beyond the position of the zero loss. These peaks are the characteristic plasmon loss peaks of the fast electrons traversing the silicon crystal. At the 100eV energy loss, additional absorption is observed and can be attributed to the onset of the silicon L-edge absorption. This demonstrates that a high quality spectrum over the energy range where the sensing is taking place has indeed been recorded. This, coupled with the ability to acquire the energy stabilized higher energy loss using the more efficient parallel detector, means that EEL spectrum of a wide dynamical energy range can be detected simultaneously.

3.4. Effect of aberration

At the moment, the ultimate energy resolution of our energy stabilized spectrometer is largely limited by the energy spread of the electron source, which is a heated tungsten filament. As the energy stabilized system is based on a post-column prism spectrometer, it is portable and can be retrofit to other electron microscope columns with a more monochromatic electron beam source. In that case, the intrinsic and parasitic aberration of the spectrometer as well as the width of the tungsten wire may be the energy resolution limiting factors. When properly tuned, for example using the above mentioned method, the spectrometer should produce a beam with a residual third or higher order aberration. This second order aberration corrected beam will interact with the wire of finite diameter to produce a registrar of the ‘no-loss’ beam position at the dispersion plane of the spectrometer, and to determine the energy resolution of the serially acquired spectrum. In the future work, our goal is to achieve higher order aberration correction so that the stabilized EELS can be acquired with a reasonably signal-to-noise ratio using a large entrance aperture.
4. Conclusion

An energy stabilization system for a post-column electron energy loss spectrometer attached to a transmission electron microscope is described and together with a new electron optical alignment method. It has been used to record the nature of energy instability and to produce an energy drift corrected electron energy loss spectrum containing the zero-loss peak. Coupled with the energy stabilized detection of higher energy losses by a parallel detector, simultaneous recording of both low and high electron energy loss over a wide dynamic range can be realized.

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