Is there a chance for mapping optical properties of buried quantum structures by means of Low Voltage Valence EELS in a STEM?

M Stöger-Pollach[1], W Hetaba[2] and S Löffler[2]

1. University Service Center for TEM (USTEM), Vienna Univ. of Technology, Vienna, Austria
2. Institute of Solid State Physics, Vienna Univ. of Technology, Vienna, Austria

stoeger@ustem.tuwien.ac.at
Keywords: valence EELS, optical properties, delocalization

The two main difficulties to overcome when performing an analysis of optical properties by employing electron beam techniques are the long-range Coulomb delocalization and the excitation of retardation losses, such as Čerenkov losses. The former problem limits the spatial resolution to a few nanometres; the latter hampers any band gap determination and the Kramers-Kronig Analysis (KKA) of the valence electron energy loss (VEELS) spectrum. In the last few years, the energy range of incident electron beams in transmission electron microscopes (TEMs) for quantitative analysis has been extended towards lower beam energies. The driving forces behind this development are manifold, like the reduction of radiation damage within the specimen [1] or the prevention of Čerenkov losses in the VEELS spectrum [2]. Some (scanning) transmission electron microscopes (S/TEMs) are operated even at 13-60 keV, still with the opportunity to employ electron energy loss spectrometry (EELS) [2]. Within this work, we discuss the advantages and limitations of conventional uncorrected S/TEMs for the determination of optical properties of buried quantum structures. We compare the spectrometric results with simulations based on density functional theory (DFT) using the WIEN2k code and with composition mapping by Z-contrast imaging and strain analysis [3].

For the low voltage STEM-EELS experiment, a conventional FEI TECNAI F20 equipped with a GATAN Tridiem energy filter was tuned for 60 kV in order to avoid relativistic losses in the VEELS spectrum. For this purpose, the extractor lens of the field emission gun and the condenser-objective lens system have to be operated in the tele-focus mode. This means that the focal lengths of the respective lenses are outside their conventional ranges. The advantage of this mode over conventional STEM settings is that the intensity in the electron beam is increased and the convergence semi-angle can be adjusted to 9.6 mrad leading to an effective spot size of 1 nm. This is close to the theoretical minimum of 0.7 nm (see Figure 1, left panel). At the same time, the diffraction disk completely falls into the spectrometer entrance aperture, thus giving the maximum signal intensity. Under these conditions, the angular range of our high angle annular dark field (HAADF) detector is 95 – 157 mrad. The inelastic delocalization for this setting is expected to be 4.7 nm, as can be seen in the left panel of Figure 1.

The investigated specimen is a conventional blue (Al,In)GaN laser diode from a DVD drive showing three Indium doped quantum wells (QWs) as depicted in Figure 1 (central panel). The sample was cut by means of a focused ion beam (FIB) in order to have a specimen of uniform thickness. A final Ar^+ ion polishing step was performed in order to eliminate any Ga contamination from the FIB preparation at the sample surfaces. The chemical composition of the InGaN QWs is analysed using EELS and Z-contrast. It is found that the Indium concentration is app. 11% within the QWs. This result is consistent with a comparison of the strain measurements in the HRTEM and HRSTEM images with [3].

Because the STEM was operated at 60 kV, Čerenkov losses need not to be considered during the data analysis [4]. The inelastic delocalization caused by the long-range Coulomb interaction between the swift electron and the specimen at a collection semi-angle of 10 mrad is 4.7 nm for 60 keV electrons. This is slightly smaller than the QW thickness of 5.2 nm. For the spectrum image (SI), a VEELS spectrum was recorded every 2.7 nm^2. The SI was subsequently corrected for multiple scattering and the zero loss peak (ZLP) was subtracted as suggested in [4]. The spectra recorded at the QWs exhibit a stronger interband transition signal at 3.4 eV compared to the ones recorded in GaN. Also, a plasmon shift towards lower energies can be observed (Figure 1, right panel). The spectra were then analysed using KKA. The normalization within KKA was performed using the constant thickness approach [5]. We found a higher refractive index in the QWs compared to the surrounding GaN, as shown in Figure 2, left panel. We also calculated the refractive index using WIEN2k employing the modified Becke-Johnson potential [6] using the chemical composition from the HAADF analysis. The difference between the measured and expected refractive indices is caused by the inelastic delocalization which is responsible for flattening the refractive index profile across the QWs (Figure 2, right panel).

We demonstrated that with standard equipment optical properties of buried quantum structures can be determined using VEELS with some limitations. If a C_s corrected STEM were used, the spatial resolution could be improved further and the determination of optical properties could be brought to the 1 nm scale.

References

[1] U Kaiser *et al*, Ultramicroscopy **111** (2011), p. 1239.
[2] M Stöger-Pollach, Micron **41** (2010) p. 577.
[3] A Rosenauer *et al*, Ultramicroscopy **111** (2011), p. 1316.
[4] M Stöger-Pollach, Micron **39** (2008) p. 1092.
[5] M Stöger-Pollach, A Laister and P Schattschneider, Ultramicroscopy **108** (2008), p. 439.
[6] F Tran and P Blaha, Phys. Rev. Lett. **102** (2009), p. 226401.
[7] The authors gratefully acknowledge funding from the Austrian Science Fund (FWF) under grant number I543-N20.

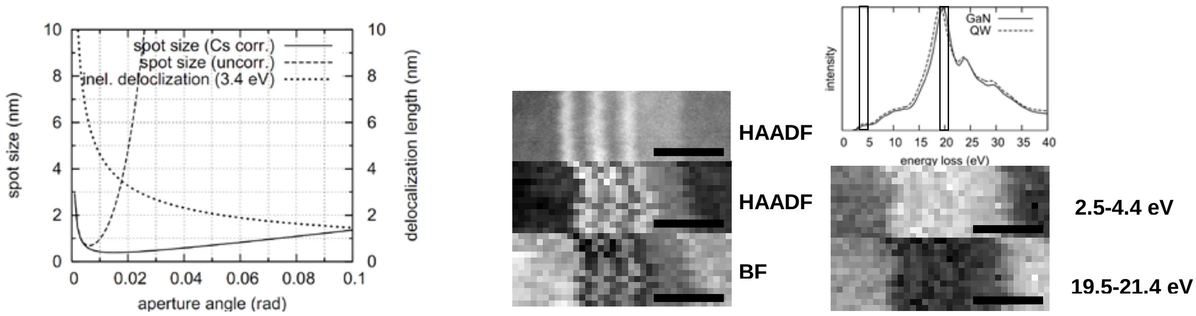

Figure 1. *Left panel: Spot size of a C_s corrected and an uncorrected 60 kV STEM vs. convergence angle and inelastic delocalization of a 3.4 eV energy loss vs. collection angle. Central panel: 60 kV HAADF STEM image of the QWs (top), HAADF STEM image recorded simultaneously with the VEELS experiment (centre) and BF STEM image extracted from the ZLP of the VEELS spectrum image (bottom). Right panel: ZLP subtracted VEELS spectra of the QW and GaN (top). The two flagged energy regions are used for inelastic imaging. Inelastic images at the interband transitions (2.5 - 4.4 eV, centre) and at the GaN plasmon (19.5 - 21.4 eV, bottom). The black scale bar represents 25 nm.*

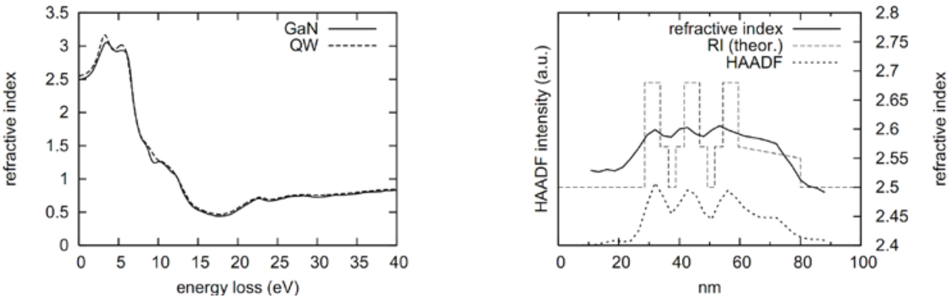

Figure 2. *Left panel: Refractive index of a QW compared to that of the GaN matrix. Right panel: Refractive index (1 eV) profile across the QWs. For better visibility, the HAADF profile and the theoretical refractive index profile are also shown. The inelastic delocalization flattens the experimental profile.*