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Optimization of experimental parameters for minimizing the inelastic delocalization in low loss EELS experiments utilizing Low-Voltage EELS

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The development of transmission electron microscopes (TEMs) has now an eighty year long tradition. In the beginning one strived to increase the beam energy in order to have better spatial resolution. With this advantage sample damage caused by the highly energetic electrons was also increased. In the last few years the development points towards lower beam energies, because lens aberrations can be corrected. This leads to excellent spatial resolution with an additional reduction of beam damage based on the knock-on mechanism. For electron energy loss spectrometry (EELS) the history of beam energies had an opposite effect: with increasing beam energies the spatial resolution was reduced because of the long range Coulomb interaction between the swift probe electron and the specimen. The actual development towards lower beam energies is therefore also advantageous for the spatial resolution of EELS experiments. Beside the spatial resolution also the inelastic cross section is increasing leading to a “stronger” EELS signal. In the present work we discuss the optimization for experimental parameters for performing EELS low loss spectrometry. The parameters which can be adjusted in order to achieve the best possible spatial resolution are (i) beam energy, (ii) convergence and (iii) collection semi-angles. Finally we present an optimized low loss experiment using a 60 keV electron beam for the determination of band gaps and optical properties of $In_{0.12}Ga_{0.88}N$ (InGaN) quantum wells (QWs) embedded in a GaN matrix.

Because the refractive index of InGaN is 2.62, the theoretical Cerenkov limit [1] is 41.843 keV. The experimental Cerenkov limit is a little bit higher, because the intensity in the relativistic losses is below the noise level as long as the normalized probability for Cerenkov photon excitation is below 0.2 - 0.4 photons per inelastic mean free path length per unit charge [1]. For the investigated materials this means that we are restricted to beam energies below 60 keV. The second aspect for the best choice of the applied beam energy is the spatial resolution in the inelastic (low loss) signal. Due to the fact that the QW thickness is 5.3 nm, the spatial resolution must be at least as good. As soon as we can avoid Cerenkov losses the lowest possible energy loss would represent the lower conduction band energy, which is 2.8 eV for InGaN [2]. In the GaN matrix the band gap is 3.24 eV. This means that the lowest observable feature in the VEELS spectrum is the 2.8 eV on-set within the InGaN QW. We therefore have to focus on the spatial resolution of the inelastic image recorded at 2.8 eV energy loss. Because the experiments are performed in scanning TEM (STEM) mode, the minimum achievable beam diameter at the respective beam energy must also be considered. Figure 1 shows the beam diameter of a FEG-STEM probe formed by a condenser lens with a spherical aberration coefficient C_S of 1.2 mm, as it is available in our TECNAI TEMs. The optimum beam diameter requires an aperture angle of 18 mrad. Then it would be equal to the inelastic delocalization of 3.8 nm, which is still below the QW width. The difference in spot size and delocalization between a 60 keV and a 40 keV electron beam is negligible. Hence only the possible appearance of Cerenkov losses determines the choice of the most appropriate beam energy. The best spatial resolution of 0.68 nm can be achieved in the 60 keV HAADF image when the convergence angle is 6 mrad. In the presented experiments we chose a convergence angle of 9 mrad, this leading to a spatial resolution of 0.8 nm. The inelastic delocalization of 5.3 nm is equal to the width of a single QW. The rectangular scan area was 22 nm x 70 nm. The effective pixel size of the STEM-EELS experiment was 4 nm^2. Figure 2b shows the HAADF image recorded simultaneously to the STEM-EELS data cube. When using the EELS data cube a ZLP filtered STEM bright filed image can be extracted (see Figure 2c). The advantage of recording a whole data cube is that single spectra can be extracted from an exactly defined region of the specimen. In Figure 2d we present two spectra, one of the GaN matrix and one of a center of a QW. It can be easily recognized that the QW shows the smaller band gap. Furthermore a plasmon shift is also visible. Finally the refractive index can be calculated for each pixel of the scan. Figure 3a shows the mean refractive index over five measurements from a QW center compared with the one from the GaN matrix. From the experimentally obtained data set a refractive index map can be generated (Figure 3b).

The main benefit of low beam energies in EELS is definitively in the energy range of optical transitions, which is the VEELS region. This is because the VEELS signal can be altered by relativistic losses and the spatial resolution suffers from the Coulomb interaction. By reducing the beam energy relativistic losses can be avoided and the spatial resolution is intrinsically improved.

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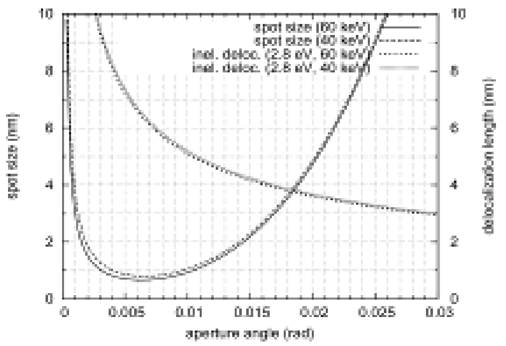

Figure1. STEM probe size as a function the convergence (aperture) angle for 60 keV and 40 keV electron probes in comparison with the inelastic delocalization for a 2.8 eV energy loss as a function of the collection aperture angle.

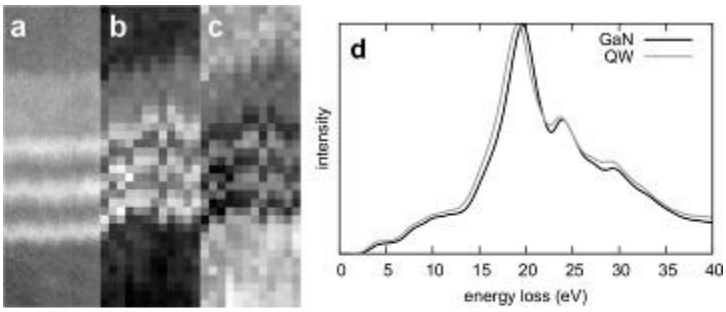

Figure 2. (a) Separately recorded 60 keV HAADF STEM image of the QWs using a short dwell time; (b) 60 keV HAADF STEM image recorded simultaneously with the STEM-EELS experiment. The pixel size of the raster scan is 4 nm^2. The scan area is 22 nm x 70 nm; (c) ZLP filtered STEM bright filed image of the respective area extracted from the STEM-EELS data cube; (d) ZLP subtracted VEELS spectra recorded in the GaN matrix and in the center of a QW. The plasmon peak shift and the smaller band gap of the QW are clearly visible.

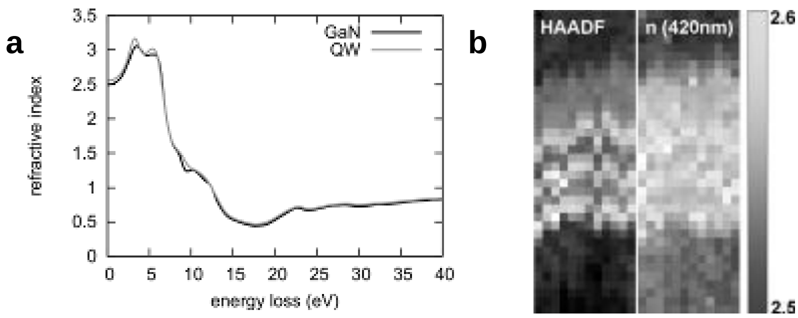

Figure 3. (a) Mean refractive indices of five GaN measurements and five measurements taken from the center of a QW; (b) Experimentally obtained HAADF image and refractive index map for an energy loss of 2.95 eV (420 nm).