Advantages of low beam energies in a TEM for valence EELS

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Abstract. Since the availability of monochromators in transmission electron microscopes (TEMs), electron energy loss spectrometry (EELS) is widely used to determine band gaps and the dielectric properties of semiconductors on a nano-metre scale. Nevertheless, three physical effects hamper straightforward analysis: (a) relativistic energy losses, (b) the delocalization of the energy loss which is in the 10 nano-metre range for valence losses, and (c) the presence of interface plasmons. When reducing the operation voltage of the TEM one can kill two birds with one stone: (a) the relativistic losses will disappear as soon as \(v < c_0/n\) (with \(v\) as the speed of the electron, \(c_0\) as the vacuum speed of light and \(n\) as the refractive index of the investigated sample) and (b) the delocalization will decrease, because it also depends on the energy of the incident electron probe. The determination of the optical properties of quantum structures is discussed in the case of GaP/GaAs interface at 200 keV and 20 keV beam energy, respectively. Further, the influence of the delocalization of the energy loss signal is discussed theoretically and experimentally.

1. Introduction

Using low beam energies in transmission electron microscopy (TEM) has a long tradition in biological samples. This has a few reasons: i) beam damage is reduced because the energy of the probe electron is reduced – and usually biological samples are very beam sensitive –, ii) due to the fact that biological samples have a large Carbon content, the mean free path for inelastic scattering is very low, this means that even samples of a thickness in the range of 70-100 nm can be easily trespassed by electrons accelerated with 80 kV and iii) when the incident electron energy is reduced and the mean free path for inelastically scattered electrons is decreased, the contrast in the image is enhanced. Beside advantages like reducing beam damage and enhancing contrast also two big disadvantages are coming up, when reducing the beam energy. First, the resolution of the microscope is reduced. This is shown in Fig. 1, where the contrast transfer function (CTF) of a TECNAI G20 microscope equipped with a super twin lens and a LaB\(_6\) electron source is plotted. In all cases the extended Scherzer defocus was selected. For biological samples the CTF seems to be still good enough, but in materials science efforts need to be done in order to bring it back to the high resolution regime. First successful results were presented on the European Microscopy Conference 2008 (EMC 2008), where a \(C_s\) corrected high resolution images recorded at 30 kV was shown [1]. The second disadvantage is the decrease of brightness when reducing the operation voltage of the TEM. Little brightness automatically means that image recording times increase and drift and other instabilities also need to be considered. Again a
way out of this dilemma was shown on the EMC 2008: a high brightness source [2]. As a consequence next generation TEMs will be able to compensate all disadvantages of lowering the high tension and give back the operator the full power of a modern transmission electron microscope.

Although such next generation machines are not available commercially at the moment, this work was done on a conventional TEM and shows that lowering the beam energy has also positive effects on valence energy loss spectrometry (VEELS). One effect has an instrumental reason: the improvement of the energy resolution. Another one was first described by N. Bohr [3] in 1913: the decrease of the delocalization of the energy loss. A third advantage is the prevention of the excitation of relativistic energy losses in semiconductors as described elsewhere [4]. Due to the fact that the third positive aspect was described in detail in [4] this work is focused on the improvement of the spectral energy resolution and on the shrinking of the delocalization.

2. Improving the spectral resolution
Conventionally, the spectral resolution is improved by using a monochromator in the condenser system of a field emission microscope (FEG-TEM). Due to the construction of conventional gun systems used in FEG-TEMs a reduction of the high tension below 60 kV reduces the intensity of the beam tremendously. With the construction of the new X-FEG, this problem is overcome.

On the other hand, the construction of a LaB₆ operated electron gun allows reducing the high tension down to 20 kV without losing too much intensity. The loss of intensity can be compensated by the Wehnelt bias. In contrast to a FEG-TEM, using a monochromator on a LaB₆-TEM is not useful, because a LaB₆ gun has much less brightness which would then be further reduced. An improvement of the energy resolution in the EELS spectrum can therefore only be obtained by a reduction of the filament temperature and an improvement of the point spread function (PSF) of the detector. Unfortunately, a reduction of the filament temperature is counterproductive, because brightness is reduced too. This means that all the improvement of the energy resolution in the loss spectrum is reached by the improvement of the PSF. Figure 2 shows the measured PSF at 200 kV and 20 kV of our GIF 2001 system. For its determination a part of

| beam energy | energy spread | ext. Scherzer defocus |
|-------------|---------------|-----------------------|
| 20 kV       | 0.35 eV       | -124.3 nm             |
| 60 kV       | 0.7 eV        | -93.6 nm              |
| 200 kV      | 1.1 eV        | -67.2 nm              |

Table 1. Instrumental conditions for the TECNAI G20 ST. Further: Cₛ = 1.2 mm, Cᶜ = 1.1 mm, dV/V = 2ppm, dI/I = 5ppm, convergence angle = 0.1 mrad

Fig. 1. Contrast transfer function of a TECNAI microscope operated at 20, 60 and 200 kV.

Fig. 2. PSF of the GIF 2001 at 200 keV and 20 keV beam energy.
the CCD was blocked in order to have a knife-edge in the energy dispersive direction. Then the CCD was illuminated homogeneously and the PSF was measured. The positive effect of the improved PSF on the energy resolution can be directly measured when the full width at half maximum (FWHM) of the zero loss peak (ZLP) is measured. Figure 3 shows the ZLPs recorded at 200 kV and 20 kV, respectively. The FWHM of the ZLP is reduced to 0.35 eV instead of 1.1 eV when the TEM is operated at 200 kV.

3. Delocalization in EELS at low beam energies

Besides an improvement of the energy resolution in the EELS spectrum, a reduction of the delocalization of the energy loss is induced when the beam energy is decreased. Figure 4 shows the reduction of delocalization at low beam energies in comparison to 100 keV and 200 keV electron beams using once Bohr’s cut-off criterion [3] and once the Rayleigh criterion [5]. The Rayleigh criterion seems to be very crude, because it does not include Coulomb interaction. The resulting values are about five times larger than those obtained by using Bohr’s cut-off criterion. Common for both models is that the delocalization is reduced by a factor of approximately 3 when using 20 keV instead of 200 keV electrons. Comparing Bohr’s cut-off criterion with a quantum mechanical model [6] shows an excellent agreement.

4. Experimental results

For the experimental determination of the optical properties of GaP and GaAs, respectively, a valence EELS (VEELS) spectrum image was recorded across the GaP/GaAs interface. This has the advantage, that the phenomena of i) delocalization and ii) interface plasmon excitation can be studied simultaneously.

4.1. Delocalization

For the VEELS experiments an FEI TECNAI G20 equipped with a GIF 2001 system was operated at 200 kV and 20 kV. The GaP/GaAs interface was rotated parallel to the energy dispersive axis of the spectrometer using a double tilt rotation holder allowing recording a \( \Delta E-r \) pattern (Figure 5, left side).

On the right hand-side of Figure 5 it can clearly be seen that for the 200 kV experiment the delocalization is in the range of 10 nm as given by Bohr’s cut-off criterion. Although the signal is much more noisy for the 20 kV experiment, the delocalization can be estimated to be roughly 4 nm, which is also in good agreement with calculations. This experimental test has a fundamental

Fig. 3. Zero loss peaks recorded with 200 keV and 20 keV beam energy.

Fig. 4. Reduction of delocalization compared to 100 keV and 200 keV electrons for an energy loss of 16.4 eV.

Fig. 5. left: Spectrum image of the GaP/GaAs interface at 200 kV and 20 kV at 16.4 eV energy loss (see arrow). right: Normalized intensity profiles at the GaP plasmon maximum for the determination of the delocalization of the energy loss.
importance for the determination of the optical properties of non-isolated quantum structures: using 200 keV electrons, a simple Kramers-Kronig Analysis of VEELS experiments is failing. Besides the influence of relativistic effects [5], delocalization also hampers VEELS analysis of nano-objects using highly energetic electrons.

4.2. Interface plasmons

Besides delocalization and relativistic energy losses a third feature is altering the VEELS signal: the interface plasmon. Due to the fact that interface plasmons (IPs) are at lower energies than volume plasmons, the delocalization of these collective oscillations is even stronger. Figure 6 (left) shows the calculated position of the IP at the GaAs/GaP interface. In the non-relativistic formalism for a flat interface between materials A and B, with the complex dielectric functions $\varepsilon_A$ and $\varepsilon_B$, the contribution of the IP is proportional to $\text{Im}[(\varepsilon_A - \varepsilon_B)/(\varepsilon_A + \varepsilon_B)]$. Strong IPs are expected when $\text{Re}(\varepsilon_A + \varepsilon_B) = 0$, and may produce loss signals below the band gap energy. For the present interface the maximum of the IP is found to be 3.4 eV (Figure 6, left hand-side). Bohr’s cut-off criterion gives a delocalization of this energy loss of 55 nm for 200 keV and still 20 nm for 20 keV probe electrons. This is shown in Figure 6 on the right hand-side.

5. Conclusion

The recent experiment demonstrated that the determination of band gaps and dielectric properties of buried quantum structures is still erroneous when using TEM-EELS applications. Although the delocalization can be decreased by decreasing the beam energy and also relativistic effect can be avoided completely, the appearance of IPs hampers straightforward analyses. Even in the case of an interface between two materials with similar dielectric functions – as the GaAs/GaP interface –, IPs can be detected.

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