Low voltage TEM for semiconductor analysis

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Abstract. Valence electron energy loss spectrometry (VEELS) is more and more employed for the quantification of dielectric properties in semiconductor research. The improvement of the energy resolution of the thermo-ionic emitter (lanthanum-hexaboride) is obtained by decreasing the acceleration-to-Wehnelt voltage ratio and by inserting a smaller Wehnelt diaphragm. These modifications lead to an improved energy resolution of 0.25 eV full width at half maximum and 3.03 eV full width at thousandth maximum at 20 keV beam energy, which is pretty close to the one of a monochromated field emission gun. Beside the improved energy resolution we demonstrate the accuracy of low voltage VEELS in terms of band gap determination of Al$_x$Ga$_{1-x}$As, with $x$ varying from zero to 0.58. The experimental results are compared with a well established semi-empirical model commonly used for the calculation of the properties of III-V semiconductors.

1. Introduction

In recent years a renaissance of low beam energies for operating transmission electron microscopes (TEMs) can be observed. The main driving force behind this development is the reduced beam damage based on the knock-on mechanism. The second benefit of employing low beam energies is the enhanced contrast in TEM and STEM imaging, due to the beam energy dependent scattering behaviour. Therefore all main TEM and scanning TEM (STEM) manufacturers apply nowadays low voltage instruments with different ranges of available beam energies. Two of these TEMs are outstanding, because they combine best imaging capabilities (by employing C$_s$-correctors) and high-resolution EELS at very low voltages. These are the JEOL microscope at AIST [1], Japan, equipped with a probe and an image C$_s$-corrector and EELS capabilities from 30 kV to 60 kV using a post column GATAN ENFINA spectrometer. The second one is the recently inaugurated ZEISS SALVE microscope [2] equipped with an image C$_s$-corrector and EELS capabilities from 20 kV to 200 kV using an in-column Ω-type energy filter. In the actual work we use a FEI TECNAI 20-200 operated at 20 kV equipped with a GATAN GIF2001 energy filter. The TECNAI 20-200 has a thermo ionic electron emitter [3,4]. Its energy resolution was improved by introducing a smaller Wehnelt aperture with a diameter of 0.3 mm thus achieving 0.6 eV full width at half maximum (FWHM) in the zero loss peak (ZLP) at 200 kV and only 0.25 eV FWHM at 20 kV operation voltage.

In the actual work, we employ low beam energies for having another benefit: we want to avoid relativistic energy losses, like Čerenkov losses and light guiding modes [5-8] which alter the low loss spectrum. Thus we are able to perform Kramers-Kronig Analyses (KKA) without the need for taking into account relativistic effects [8,9]. From KKA we gain the energy loss function and further the optical constants, like refractive index, absorption coefficient, band gap, complex dielectric function –

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and all of them for a much broader energy range than would be measurable by employing optical methods. So the advantage of electron beam techniques is clear: the spectral range and the spatial resolution are much better compared to optical methods. The latter benefit is limited by the Coulomb delocalization [4], which is in the range of several nanometres for very low losses. However, the delocalization is also reduced by using lower beam energies.

2. Experimental

The specimen was a layered structure of two $\text{Al}_x\text{Ga}_{1-x}\text{As}$ layers on a GaAs substrate. The chemical composition was characterized by means of energy dispersive X-ray analysis (EDX). Table 1 gives the chemical compositions of the layers. The layer thickness was 200 nm for layer 1 and 400 nm for layer 2. The sample was prepared by using a focused ion beam. A final Ar$^+$-ion polishing step was performed. The final lamella thickness in the investigated area varied from 77 nm – 82 nm.

The valence EELS (VEELS) experiments were performed on a conventional FEI TECNAI 20-200 operated at 20 kV. The measured energy resolution was 0.25 eV in the FWHM of the ZLP using a fully saturated filament (see Figure 1). The full width at thousandth maximum is 3.03 eV, which is the more important value for band gap studies compared to the FWHM.

Due to the fact that Čerenkov losses are strong in GaAs [6,7] and also in $\text{Al}_x\text{Ga}_{1-x}\text{As}$, the beam energy was reduced such, that any relativistic energy losses were avoided experimentally. This was achieved by using a 20 keV electron beam. The microscope itself was operated in image mode, because the smallest available selected area aperture is 260nm back-projected onto the sample. For the thinnest layer (layer 1) this would be too large by a factor of two. For the VEELS experiments the collection semi-angle was kept small, in order to be able to compare the results with optical data, which are intrinsically limited to zero momentum transfer. The selected collection semi-angle was 2.8 mrad, which is due to the smallest available objective aperture (OA). Table 2 gives the relative size of the smallest OA in comparison with the dimensions of the Brillouin zone of GaAs for 20 kV. However, the strong band bending of GaAs must be considered when comparing the VEELS results with optical data.

3. Results on $\text{Al}_x\text{Ga}_{1-x}\text{As}$

In the following we compare the experimental results by the theoretical model established by Adachi [10,11], where the refractive index $n(\omega)$ in zincblende materials below the direct band gap edge can be expressed as

$$n(\omega) = \sqrt{A_0 \cdot (f(\chi) + 0.5 \cdot \left( \frac{E_0}{E_0 + \Delta_0} \right)^{3/2} \cdot f(\chi_{SO}) + B_0}}$$

| location | distance\(^a\) [k] | $\beta$ [mrad]\(^b\) |
|----------|-----------------|-------------------|
| $\gamma = 000$ | - | - |
| $\chi = 100$ | $|\gamma \cdot \chi| = 1$ | 0.373 |
| $L = \frac{111}{2} \frac{1}{2}$ | $|\gamma \cdot L| = \sqrt{2}$ | 0.431 |
| $K = \frac{330}{4} \frac{1}{4}$ | $|\gamma \cdot K| = \sqrt{5}$ | 0.352 |

*Distances in dimensionless “q-space” units.

\(^b\) $\beta = 2.83$ mrad, $E_0 = 20$ kV

Table 2: Relative size of the collection aperture $\beta$ (2.8 mrad) to the Brillouin zone of GaAs for 20 kV.
with

\[
f(\chi) = \frac{2 - \sqrt{1 + \chi} - \sqrt{1 - \chi}}{\chi^2}
\]

\[
\chi(\omega) = \frac{\omega}{E_0}
\]

\[
\chi_{SO}(\omega) = \frac{\omega}{E_0 + \Delta_0}
\]

where \(A_0, B_0, E_0,\) and \(\Delta_0\) are given in [11]. \(\Delta_0\) represents the spin-orbit coupling energy, \(E_0\) is the fundamental band gap at the \(\Gamma\)-point – which is plotted as a function of the Al concentration \(x\) in Figure 2 (left). This figure also shows the energy separation in the \(X\) and \(L\) point of the Brillouin zone. As long as the \(\Gamma\)-line (full line) is the lowest one, \(\text{Al}_x\text{Ga}_{1-x}\text{As}\) has a direct band gap, but as soon as the \(X\)-line (dashed line) has crossed, \(\text{Al}_x\text{Ga}_{1-x}\text{As}\) has an indirect gap. The right hand side of Figure 2 shows the experimentally obtained band gaps of the three layers.

**Figure 2**: Left: separation energies in the \(\Gamma, X\) and \(L\) points of the Brillouin zone as a function of the Al concentration \(x\) in \(\text{Al}_x\text{Ga}_{1-x}\text{As}\). Right: Plural scattering deconvolved 20 kV VEELS spectra of \(\text{Al}_x\text{Ga}_{1-x}\text{As}\) with \(x=0\) (full line), \(x=0.5\) (dashed line) and \(x=0.58\) (dotted line). The insertion shows the single scattering distributions in the energy range from 1-5 eV.

**Figure 3**: Left: Refractive index calculated after [11]. Right: Refractive index obtained via KKA from the experimental VEELS results of \(\text{Al}_x\text{Ga}_{1-x}\text{As}\). With \(x=0\) (full line), \(x=0.5\) (dashed line) and \(x=0.58\) (dotted line).
3.1. **Kramers-Kronig Analysis (KKA)**
Before KKA the experimental spectra were deconvolved from plural scattering using the matrix approach [12]. Further the surface loss contributions were removed from the single scattering distribution following Kröger’s fully relativistic theory [5] and the spectra were normalized using the sample thickness approach [8]. The results fit very well the calculated ones. On the left hand side of Figure 3 the calculated values of the refractive index for the band gap [11] are displayed and on the right hand side the experimental obtained values for a much larger energy range are shown. As displayed in Figure 3 (right) the refractive index fits also very well the calculated data.

3.2. **Band gaps**
Due to the small collection semi angle the indirect band gaps cannot be observed. The measured band gaps therefore follow the $\Gamma$-line in Figure 2. In Figure 4 the experimental (bullets with error bars) and calculated (full line) band gaps are displayed. The uncertainty stems first from the spectral broadening due to the initial energy width of the electron beam and second from the signal to noise ratio (SNR). Due to the fact that in the band gap region the signal is very low, the SNR is worse than in the rest of the VEELS spectrum. The indirect band gap was outside the collection aperture and can therefore not be measured.

4. **Discussion**
In this work we exemplarily showed the accuracy of the determination of dielectric properties by employing VEELS. Under the condition that relativistic effects can be avoided VEELS is a reliable tool. Due to the fact that a small collection semi angle is chosen for having a better comparability with optical methods, indirect band gaps cannot be determined. However, the direct transitions are measured very accurately. For measuring indirect band gaps in Al$_x$Ga$_{1-x}$As the collection aperture must be located at the $X$-point of the first Brillouin zone. This kind of dark field VEELS was done for Silicon in [13]. Because Čerenkov losses and light guided modes induce strong forward scattering ($\theta_{\text{Čerenkov}} < 200$ $\mu$rad), their signature cannot be recorded in dark field VEELS thus allowing higher beam energies. However, such results are not comparable with the ones from optical methods.

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