Ultramicroscopy 109 (2009) 1183–1188

Contents lists available at ScienceDirect

Ultramicroscopy

journal homepage: www.elsevier.com/locate/ultramic

Bandgap measurement of thin dielectric films using monochromated STEM-EELS

Jucheol Park *, Sung Heo, Jae-Gwan Chung, Heekoo Kim, HyungIk Lee, Kihong Kim, Gyeong-Su Park

Samsung Advanced Institute of Technology, AE Center, Nongseo-Dong Mt. 14-1, Giheung-gu, Yongin-si, Gyeonggi-do 446-712, Republic of Korea

ARTICLE INFO

Article history:
Received 17 June 2008
Received in revised form
6 April 2009
Accepted 28 April 2009

PACS:
82.80.Dx
79.20.Uv
68.37.Ma
77.55.+f

Keywords:
Bandgap measurement
Monochromtor
STEM
EELS
AES
REELS
a-SiNx
SiO_2
Charge trap memory

ABSTRACT

High-resolution electron energy-loss spectroscopy (HR-EELS), achieved by attaching electron monochromators to transmission electron microscopes (TEM), has proved to be a powerful tool for measuring bandgaps. However, the method itself is still uncertain, due to Cerenkov loss and surface effects that can potentially influence the quality of EELS data. In the present study, we achieved an energy resolution of about 0.13 eV at 0.1 s, with a spatial resolution of a few nanometers, using a monochromated STEM-EELS technique. We also assessed various methods of bandgap measurement for a-SiNx and SiO_2 thin dielectric films. It was found that the linear fit method was more reliable than the onset reading method in avoiding the effects of Cerenkov loss and specimen thickness. The bandgap of the SiO_2 was estimated to be 8.95 eV, and those of a-SiNx with N/Si ratios of 1.46, 1.20 and 0.92 were measured as 5.3, 4.1 and 2.9 eV, respectively. These bandgap-measurement results using monochromated STEM-EELS were compared with those using Auger electron spectroscopy (AES)-reflective EELS (REELS).

1. Introduction

The bandgap measurement of thin dielectric films is essential to improving the reliability of charge trap flash memory, which is accomplished by bandgap engineering of the silicon nitride and oxide layers used as traps and tunneling layers, respectively [1]. Traditionally, the bandgaps of dielectrics have been measured by optical methods offering high-energy resolution (~ several tens of meV) but very poor spatial resolution (~0.2 μm). This spatial resolution clearly is insufficient for measuring bandgaps in modern devices with horizontal and vertical structures in the nanometer range. Hence, there has been an increasing demand for both high-energy and high-spatial-resolution techniques for bandgap measurement.

Recently, high-resolution electron energy-loss spectroscopy (HR-EELS), achieved by attaching electron monochromators to transmission electron microscopes (TEM), has proved to be a powerful tool for bandgap measurements on the nano-scale [2–8]. Despite this technological breakthrough, however, there remain restrictions for spatial resolution and precision of measurement on monochromated TEM. First, the ultimate spatial resolution obtainable by TEM is limited by the beam size and the delocalization phenomenon. Second, estimation of precise bandgaps in the STEM mode, which offers superior spatial resolution, is still uncertain, due to Cerenkov loss [9–12] and various effects that impose artifacts on bandgap measurements for a wide range of thin dielectric films [13,26].

In this paper, several methods of TEM bandgap measurement for thin dielectric films were assessed by means of comparisons with the Auger electron spectroscopy-reflective EELS (AES-REELS) method. We report that we measured reliable bandgap values of thin dielectric films using the linear fit method in STEM mode with an energy resolution of about 0.13 eV at 0.1 s and a spatial resolution of several nanometers using our monochromated Schottky-FEG TEM.

2. Experiments

SiNx films from various SiH_4:NH_3:N_2 gas mixtures at 400 °C were deposited at 670 W RF power by the plasma-enhanced

* Corresponding author. Tel.: +82 31 280 9141; fax: +82 31 280 9157.
E-mail address: jucheol.park@samsung.com (J. Park).

0304-3991/$ - see front matter © 2009 Elsevier B.V. All rights reserved.
doi:10.1016/j.ultramic.2009.04.005

chemical vapor deposition (PECVD) method on (0 0 1) p-Si. The SiO_2 films were grown by conventional thermal oxidation. TEM specimens were prepared according to the mechanical-thinning and ion-milling method. Plasma cleaning was carried out before the TEM work in order to acquire the bandgap spectra.

The STEM-EELS spectra were obtained using Field-emission TEM (FEI Titan 80-300) equipped with a Wien-type monochromator and a high-resolution Gatan imaging filter (GIF Tridiem 865 ER300) installed at the Samsung Advanced Institute of Technology. The acceleration voltage of the TEM was 300 kV. The energy spread of a monochromated zero-loss peak (ZLP) was 0.13–0.27 eV in full-width at half-maximum (FWHM), depending mainly on energy dispersion, sample thickness and monochromator alignment. The energy dispersion was 0.01 eV/ch for the energy resolution and 0.02 eV/ch for the bandgap. The collection angle was 5–10 mrad, which varied with camera length (< 100 mm) and the entrance aperture size of the HR-GIF (1.0 or 2.5 mm in diameter). The EELS spectra for various thicknesses (50–200 nm) were acquired in the STEM imaging mode.

In order to compare the bandgaps estimated by STEM-EELS, AES-REELS spectra were measured on a VG Microlab 350F with primary electron energies of 1000 eV in the constant analyzer energy mode of 10 eV pass energy. The FWHM of the elastic peak was 0.8 eV. The energy-loss range was 0–100 eV.

For accurate composition analysis of Si and N, an HR Rutherford backscattering system equipped with a magnetic sector analyzer capable of obtaining a high-resolution energy spectrum of the MEIS level (KOBELCO HRBS-V500) was used. The RBS analysis was carried out with a 400 keV He^{+} probe beam, which was incident at the angle of 50° with a scattering angle of 70.5°. In order to avoid surface damage, the measurement points for the RBS were continually changed.

3. Results and discussion

3.1. Energy resolution of monochromated EELS

The energy resolution of an entire TEM-EELS system is one of the most important parameters in EELS analysis. There are four main factors associated with practical energy resolution in EELS: (a) the energy spread of the electron source, (b) the non-isochromaticity of the spectrometer, (c) the point spread (blurring) of the detector, and (d) instabilities, such as stray magnetic fields, in the TEM-high-voltage and room environments. A monochromator with a high-resolution spectrometer and a high-voltage tank directly reduces the energy spread of an electron beam, naturally resulting in an attainable high-energy resolution.

In general, the FWHM of ZLPs is regarded as an energy-resolution indicator. Fig. 1(a) shows the ZLPs acquired from a normal Schottky-FEG TEM and those derived with a monochromated Schottky-FEG TEM. TEM equipped with a thermally assisted Schottky field emission source typically produces, under normal operating conditions, an energy resolution of about 0.7 eV and an asymmetrical ZLP, which complicates the removal of the ZLP in measuring the bandgap. By contrast, monochromated Schottky-FEG TEM equipped with a Wien-type monochromator, high-resolution electron spectrometer and a high-resolution high-tension tank offers an energy resolution of about 0.13 eV and a relatively symmetrical ZLP (Fig. 1(a)).

Fig. 1(b) shows, on a logarithmic scale, the normalized intensities of the ZLPs acquired from the normal and monochromated Schottky-FEG. When the normalized intensities are shown on a logarithmic scale, the tail shape and range of the intensities are much enhanced, allowing us easily to recognize the

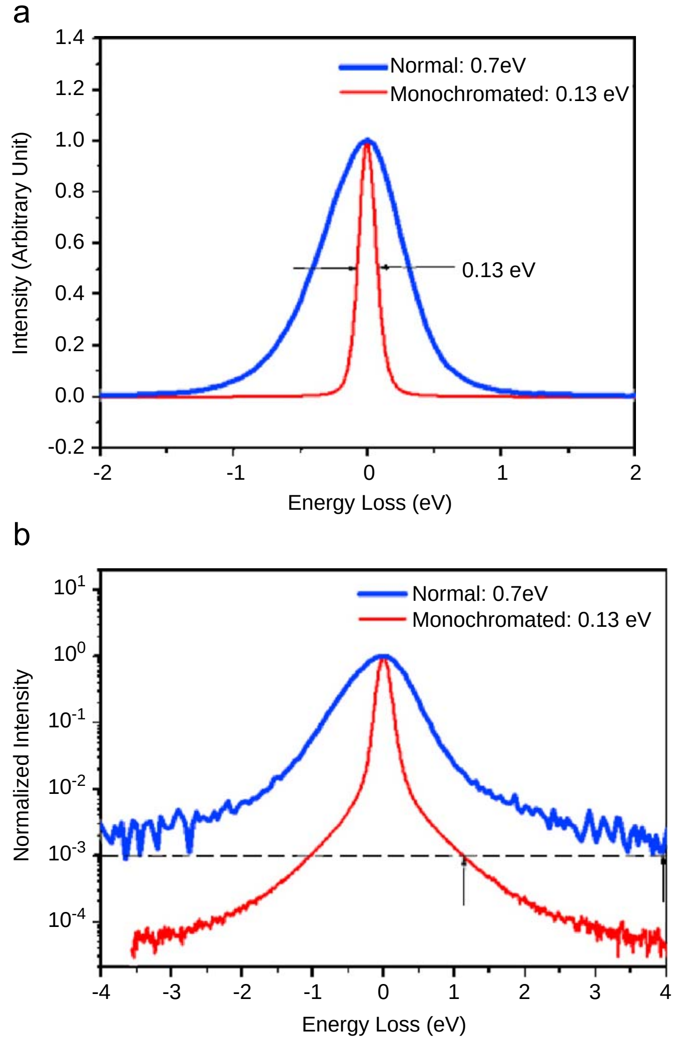

Fig. 1. Zero loss peaks acquired from the conventional and the monochromated Schottky-FEG. Normalized intensities are shown on (a) a linear scale and (b) a logarithmic scale. The full-widths at half-maximum of the conventional and the monochromated Schottky-FEG are 0.7 and 0.13 eV, respectively. Two arrows at 1.1 and 3.9 eV show the energy-loss values at a 1/1000th maximum of the conventional and the monochromated Schottky-FEG, respectively.

noise and background levels. The FWHM of the monochromated and conventional Schottky-FEGs were 0.13 eV and 0.7, respectively. The background intensities in the region of interest (1–10 eV) for a monochromated Schottky-FEG are considerably lower than those for a normal Schottky-FEG. Thus, if the threshold for bandgap detection is set at 1/1000th of the maximum intensity, as suggested in the literature [14,15], a horizontal line can be drawn at this level, so that its intersection point can be assigned with the tails of the zero-loss distribution as the minimum energy loss that can be detected above the background. The lowest detection limit for a dispersion of 0.01 eV was 1.1 eV for the monochromated ZLP and ~4.0 eV for the normal schottky-FEG. It should be noted that the difference in the minimum energy between a conventional and a monochromated schottky-FEG is substantial and critical to the accurate determination of bandgap energy.

3.2. Bandgap measurement of SiO_2

The bandgap refers to the energy difference between the top of the valence band and the bottom of the conduction band. Fast