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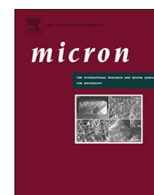

Determination of atomic-scale chemical composition at semiconductor heteroepitaxial interfaces by high-resolution transmission electron microscopy

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The determination of atomic structures and further quantitative information such as chemical compositions at atomic scale for semiconductor defects or heteroepitaxial interfaces can provide direct evidence to understand their formation, modification, and/or effects on the properties of semiconductor films. The commonly used method, high-resolution transmission electron microscopy (HRTEM), suffers from difficulty in acquiring images that correctly show the crystal structure at atomic resolution, because of the limitation in microscope resolution or deviation from the Scherzer-defocus conditions. In this study, an image processing method, image deconvolution, was used to achieve atomic-resolution (~1.0 Å) structure images of small lattice-mismatch (~1.0%) AlN/6*H*-SiC (0001) and large lattice-mismatch (~8.5%) AlSb/GaAs (001) heteroepitaxial interfaces using simulated HRTEM images of a conventional 300-kV field-emission-gun transmission electron microscope under non-Scherzer-defocus conditions. Then, atomic-scale chemical compositions at the interface were determined for the atomic intermixing and Lomer dislocation with an atomic step by analyzing the deconvoluted image contrast. Furthermore, the effect of dynamical scattering on contrast analysis was also evaluated for differently weighted atomic columns in the compositions.

1. Introduction

Heteroepitaxial films are the most widely used form of semiconductor materials. The heteroepitaxial interfaces, often as the source of misfit defects and the enrichment region of impurities and different phases, significantly affect the quality of films. Usually, active heteroepitaxial interfaces show physical or chemical modifications at atomic scale, such as atomic displacements (Jia et al., 2011; He et al., 2012), change in component concentrations (i.e., atomic occupancy) (Jia and Urban, 2004; Yang et al., 2012), or variation in chemical compositions (Schulz et al., 2012). Because of the simplicity of semiconductor crystal structure, high-resolution transmission electron microscopy (HRTEM) is commonly used to study heteroepitaxial interfaces (Otsuka et al., 1986; Nutt et al., 1987; Vila et al., 1996; Wang et al., 2011). However, these studies are mainly focused on the interface structure or the effects of interface type on the macroscopic properties of semiconductor films; more detailed studies on physical or chemical modifications at atomic scale are still lacking, causing ambiguity in the interface characteristics and their function. An important reason is that interfacial lattice distortion is usually severe, thus requiring HRTEM images with a very high resolution, even higher than that for a perfect film region, to obtain atomic-scale quantitative information.

With the development of HRTEM, aberration-corrected TEM can achieve atomic resolution by reducing the third-order spherical-aberration (C_S) coefficients of the objective lens to zero or even negative values. The microscope point resolution under the Scherzer-defocus (Δf_{Sch}) (Scherzer, 1949) (or the optimum-defocus, Δf_{opt} (Lentzen et al., 2002)) conditions is significantly improved and close to the microscope information limit, even reaching subangstrom resolution. Furthermore, the negative C_S imaging model can increase the signal-to-noise ratio of image (Jia et al., 2010). Then, the atomic-scale quantitative information is obtained by strictly comparing the experimental images with a series of simulated images with different quantitative information and imaging parameters. For example, the displacement of Zr-Ti atoms in $PbZr_{0.2}Ti_{0.8}O_3$ domain (Jia et al., 2011), displacement of both Zn and O atoms in the fresh $(10\bar{1}0)$ facets of ZnO nanosized islands (He et al., 2012), concentration of oxygen in Σ3{111} twin boundaries of $BaTiO_3$ thin films (Jia and Urban, 2004), local oxygen occupancy in $LuFe_2O_{4-\delta}$ (Yang et al., 2012), occupancy of Al cations in the partial dislocations of deformed α-Al_2O_3 (Heuer et al., 2010), and In distribution within a step

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graded $In_xGa_{1-x}N$ quantum well (Schulz et al., 2012).

However, this method to achieve atomic-scale quantitative information by directly using experimental aberration-corrected HRTEM images still has some limitations: (1) The experimental images must be acquired near Δf_{Sch} (or Δf_{opt}). If the imaging conditions deviate from them, the aberration-corrected HRTEM images, same as the conventional HRTEM images, will be subject to contrast transfer function (CTF) modulation and exhibit distortion, resulting in images that do not correctly reflect the crystal structures. (2) At Δf_{Sch} (or Δf_{opt}), the phase contrast in the low and high frequency regions will be subjected to severe attenuation, and thus image artifacts may appear, which are not beneficial for ultrahigh-resolution structure details (Lentzen, 2008). (3) The corrected state for aberration-corrected TEM is intrinsically unstable, particularly when acquiring images at resolutions near the microscope information limit (Schramm et al., 2012). (4) Even if the lens aberrations are corrected, still a certain degree of residual aberrations may exist, increasing the image delocalization and thus decreasing the image resolution (Lentzen, 2008). Similarly, although aberration-corrected scanning TEM (STEM) images can also achieve the same level of resolution as the aberration-corrected HRTEM images, the STEM image contrast gradually worsens with the increase in specimen thickness (t), especially in the defect and heteroepitaxial interface regions. Furthermore, the defect or the epitaxial interface regions are easily contaminated or damaged when acquiring STEM images, because the electron beam is converged in these areas.

Another method to obtain atomic-scale quantitative information is using ordinary conventional field-emission-gun (FEG) or aberration-corrected HRTEM images in combination with image processing. The HRTEM image processing techniques mainly include exit-wave reconstruction (Schiske, 1968; Coene et al., 1992; Kirkland et al., 1995) and image deconvolution (Li and Fan, 1979; Han et al., 1986; Hu and Li, 1991), two categories. Although the approaches are different, the purpose of image processing methods is the same: to eliminate the image distortion caused by CTF. The achieved resolution after image processing is better than the microscope point resolution and can be close to the microscope information limit. Especially, the deconvolution method to solve the inverse problem in HRTEM, skipping the exit wave, goes directly from the image to the structure. Using the deconvolution method, an ordinary single HRTEM image obtained using a conventional medium-voltage (200 and 300 kV) FEG TEM, which costs much lower than an aberration-corrected TEM, can be restored to the structural image with the resolution close to 1.4–1.0 Å (depending on the microscope information limits) (Wang et al., 2002; Tang and Li, 2005). These restored structure images (a structure image can intuitively reveal the projected structure of examined sample) are enough to distinguish the atomic-scale structure for most semiconductor materials. Furthermore, the deconvolution method can effectively avoid the abovementioned limitations of directly using experimental aberration-corrected HRTEM images (Chen et al., 2004; Tang et al., 2006; Wen et al., 2015). First, it is possible to broaden the experimental imaging conditions (no need for Δf_{Sch} or Δf_{opt}) to obtain structure images using aberration-corrected TEM. Furthermore, because the aberration-corrected HRTEM images are not taken near Δf_{Sch} (or $\Delta fopt$), which are usually near Gaussian focus for small C_S, it can avoid the severe attenuation of phase contrast in the low and high frequency regions, and then the image processing is used to restore the experimental image to the structure image with a high signal-to-noise ratio.

However, most of these studies focused on only microstructure determination. Only a few studies have been reported on the use of image processing methods to obtain atomic-scale quantitative information, for example, atomic displacements and occupancies of the Cu–O chains in the 90° grain boundary of the $YBa_2Cu_3O_{7-\delta}$ film (Houben et al., 2006), oxygen atomic occupancy at a $SrTiO_3$ dislocation core (Jia et al., 2005), variations in the cationic occupancy of K_xWO_3 (Kirkland et al., 2007), number of atoms in the atom column of Au [110] (Wang et al., 2012), and composition analysis at the InGaSb-on-InAs interface (Mahalingam et al., 2006). Recently, using the aberration-corrected HRTEM image combined with deconvolution processing and geometrical phase analysis (GPA), the strain distribution, reflecting the atomic displacements, of misfit dislocation cores at AlSb/GaAs interface was measured, and a relationship was observed with the atomic structure of dislocation cores (Wen, 2017). The strain distribution measurement using deconvoluted images in this study should be more accurate than that using experimental aberration-corrected HRTEM images in the study of stacking faults near the AlSb/GaAs interface (Wen et al., 2014). Furthermore, the contrast in structure images, restored by the deconvolution processing of aberration-corrected HRTEM images, for a 3*C*-SiC specimen with changes in C (or Si) atomic occupancy and thickness has also been studied (Wen and Smith, 2016). Besides the atomic displacement and change in atomic occupancy, the variation in atomic compositions is more frequently observed at the active semiconductor heteroepitaxial interfaces. Therefore, in this study, AlN/6*H*-SiC (0001) and AlSb/GaAs (001) interfaces were used as examples to demonstrate the image deconvolution could help improve the resolution of HRTEM images and obtain atomic-scale quantitative information. Variations in the atomic-scale chemical composition at AlN/6*H*-SiC and AlSb/GaAs interfaces were determined from simulated conventional 300-kV FEG HRTEM images in combination with deconvolution processing.

2. Interfacial chemical composition models and HRTEM image simulation

III-V family semiconductors have been widely used in high-temperature, high-frequency, and high-power electronic devices, low-power, low-noise, and high-speed devices, and infrared light-emitting devices. Their heteroepitaxial films commonly selected 6*H*-SiC, sapphire (α-Al_2O_3), or semi-insulating GaAs as the substrates. Therefore, different types of heteroepitaxial interfaces are formed between III–V semiconductor films and these substrates, such as small lattice-mismatch interfaces (AlN/6*H*-SiC, AlAs/GaAs, etc.) and large lattice-mismatch interfaces (AlSb/GaAs, GaN/Al_2O_3, etc.). Furthermore, three possible interface regions are usually observed, i.e., the flat region, stepped region, and defect region. The determination of chemical compositions in these interfaces is significant to determine the polarity of heteroepitaxial films (Ponce et al., 1996a) or formation mechanism of misfit dislocations (He et al., 2011).

Among these interfaces, AlN/6*H*-SiC (0001) interface is expected to be highly coherent because of the similarities in lattice parameters and thermal expansion coefficients between AlN and SiC basal planes. The atomic structure of AlN/6*H*-SiC (0001) interface has been studied by some research groups (Tanaka et al., 1995; Ponce et al., 1996a). Tanaka et al. compared the structure of AlN/6*H*-SiC interface between films grown on vicinal and on-axis SiC surfaces based on the observation of HRTEM images (Tanaka et al., 1995). Then, they suggested that the presence of 6*H*-SiC steps on the vicinal SiC surfaces introduces planar defects at the initial stage of film growth. Ponce et al. further proposed possible chemical bonding configurations of AlN/6*H*-SiC interface (Ponce et al., 1996a,1996b), i.e., two intermixed configurations of Si—N and Al—C bonds based on the analysis of its conventional HRTEM image characteristics and requirement of interfacial charge neutrality. A similar approach was also applied to analyze the possible bonding configurations of *m*-plane AlN/6*H*-SiC (Zhou et al., 2009), GaN/6*H*-SiC (Stirman et al., 2010), and InAs/$Ga_{1-x}In_xSb$ interfaces (Quan et al., 2010). However, there is still no direct experimental evidence or method to prove these models. Furthermore, for a large lattice-mismatch AlSb/GaAs interface, the atomic steps in Lomer dislocation have been identified, but their atomic configurations (or compositions) were just speculated according to the GPA results and simulated HRTEM images. Therefore, neither AlN/6*H*-SiC nor AlSb/GaAs interface clearly provides the atomic-scale chemical compositions, including methods to determine them.

The [11$\bar{2}$0] and [110] projected structure models of AlN/6*H*-SiC

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