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Atomic-scale 3D reconstruction of antiphase boundaries in GaP on (001) silicon by STEM

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ABSTRACT

In order to overcome the limitations of silicon-based electronics, the integration of optically active III–V compounds is a promising approach. Nonetheless, their integration is far from trivial and control as well as understanding of corresponding growth kinetics, and in particular the occurrence and termination of antiphase defects, is of great relevance. In this work, we focus on the three-dimensional reconstruction of such boundaries in gallium phosphide from single scanning transmission electron microscopy images. In the high angle annular dark-field imaging mode, the appearance of these antiphase boundaries is strongly determined by the chemical composition of each atomic column and reflects the ratio of transmitted anti- to mainphase. Therefore it is possible to translate measured intensities to the depth location of these boundaries by utilizing simulation data. The necessary spatial resolution for these column-by-column mappings is achieved via electron optical aberration correction within the microscope. Hence, the complete 3D orientation of these defects can be measured at atomic resolution and correlated to growth parameters. Finally, we present a method to reconstruct large areas from well sampled images and retrieve information about complex embedded nanoscale structures at the atomic scale.

1. Introduction

Silicon-based semiconductor devices are extremely wide spread and – even up to now – almost inevitable when thinking of the near-future development in computer technology (Wang et al., 2017). The research is branched into several different technologically relevant directions ranging from improved channel materials for transistors to the direct monolithic integration of III–V lasers on silicon for data transmission (del Alamo, 2011; Desplanque et al., 2012; Liebich et al., 2011).

Nonetheless, it is well known that the monolithic integration of III–V materials into silicon is challenging and antiphase domains (APDs) can be formed during heteroepitaxy of polar materials on nonpolar layers due to monoatomic surface steps. Although the reduction and suppression of APDs for the interesting and equally important gallium phosphide (GaP) on silicon has seen great progress, their occurrence is still a severe issue and can affect the performance of many devices adversely due to local strain and charge effects (Beyer et al., 2012, 2011; Desplanque et al., 2012; Feifel et al., 2017; Kroemer, 1987; Kunert et al., 2008; Németh et al., 2008; Volz et al., 2011). Therefore, a detailed understanding of their formation, appearance and ultimately termination is of great relevance to the integration of III–V optoelectronic materials onto exact silicon as it is used in the current complementary metal–oxide–semiconductor (CMOS) based technology.

Up to now, the detailed bonding characteristics within such antiphase boundaries (APBs) are not well understood. Although theoretical investigations have been carried out for a limited number of interfaces like GaAs/Ge and GaP/Si, experimental atomic resolution studies by Beyer et al. show the occurrence of more complex geometries than initially anticipated (Beyer et al., 2013; Cho and Carter, 2001; Rubel and Baranovskii, 2009). Due to this complex situation, the effects of APBs on many devices need careful modelling for example by atomistic simulations like density functional theory (DFT) where the resulting charge distribution is dependent on the actual geometry. Hence, the actual geometry of these APBs is important due to the charge accumulated by wrong bonds.

In order to resolve the structure of APDs with atomic resolution we use quantitative high angle annular dark-field (HAADF) scanning transmission electron microscopy (STEM). While other techniques like (S)TEM (tilt-series) electron tomography (Midgley, 2003; Midgley and Weyland, 2003) or atomic probe tomography (APT) (Kelly and Miller, 2007; Seidman, 2007) provide a reasonable resolution with an accuracy of up to 1 nm (and even below for APT), the lateral resolution of aberration corrected STEM is in the sub-Ångstrom regime. This is in particular aiding the determination of complex binding geometries as

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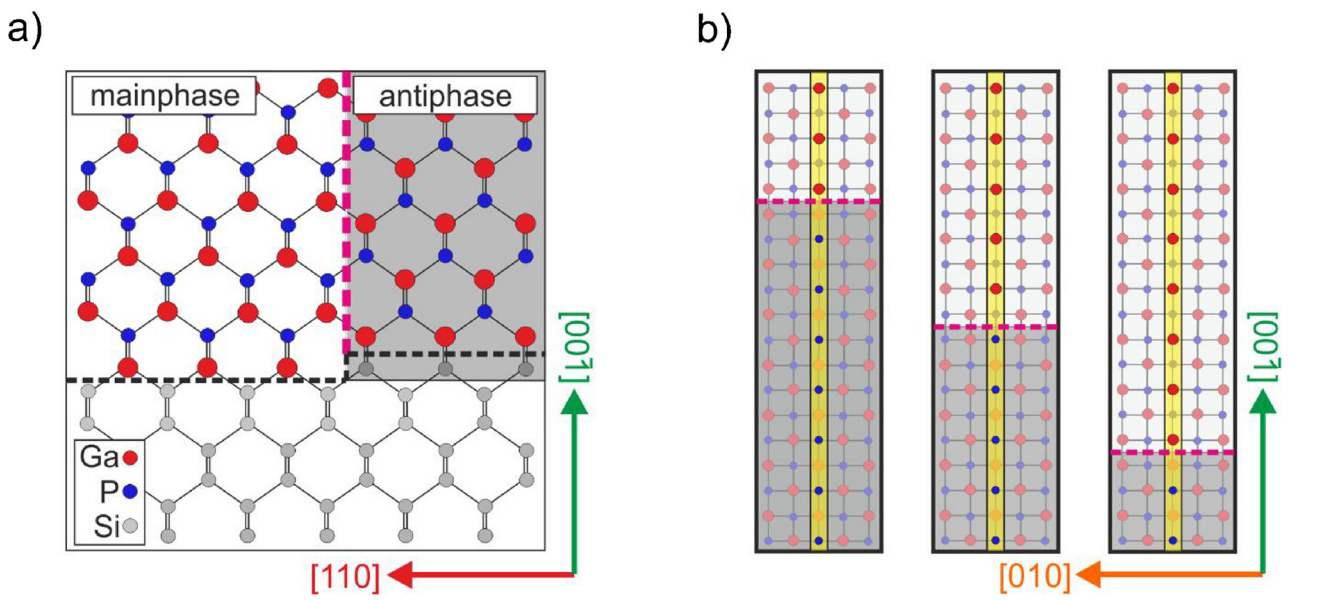

Fig. 1. (a) Cross-section view illustrating the origin of the antiphase defect (simplified) seen along [−110]. (b) The APB (magenta) divides the MP from the AP resulting in a varying ratio of Ga to P in each column.

seen for extended structural defects like APBs.

Another common disadvantage of many tomography methods is that the reconstruction relies on the acquisition of multiple images taken under different conditions and usually implies a difficult sample preparation. The latter point makes these tomography techniques impractical if either the region of interest is not representing the majority of the material or hard to find during the process of preparation.

On the other hand, quantitative STEM investigations have come a long way since the term “Z-Contrast” was coined in the early 90s of the last century (Pennycook and Jesson, 1991). The advance of instruments in the field of electron microscopy, especially the advent of commercialized aberration correctors for TEM and STEM, opened the doors for quantitative sub-Ångstrom microscopy on a daily basis (Haider et al., 1998; Krivanek et al., 1997; Uhlemann and Haider, 1998). Since then, quantifying the HAADF signal on an absolute scale at atomic resolution has become increasingly popular.

While there are many occasions where STEM measurements with qualitative match to simulations are already sufficient, the demand for quantitative match to simulations has led to a more detailed understanding of (S)TEM contrast in terms of thermal diffuse scattering (Howie, 2004; Loane et al., 1991), inelastic scattering (Allen et al., 2015; Bleloch et al., 1994; Mkhoyan et al., 2008), as well as the development of various implementations of image simulation algorithms like the STEMsalabim code (Oelerich et al., 2017) used in this work. A comprehensive list of image simulation software can be found in (Oelerich et al., 2017). In addition, the characterization of experimental equipment has led to remarkable agreement between experiment and simulation (Beyer et al., 2016a; Jones et al., 2014; LeBeau et al., 2008, 2010; Martinez et al., 2014). Especially the confocal nature of the STEM technique has been shown to truly deliver 3D information on a nearly atomic scale (Alania et al., 2016; Gonnissen et al., 2016; Muller et al., 2004). Nonetheless, even single STEM HAADF images can be used on their own to retrieve three-dimensional data in many cases as will be shown in this manuscript.

Since STEM images are generally projections of 3D entities, a direct interpretation of such images demands specific knowledge about the sample under investigation. Conventionally, in analytical STEM measurements local material compositions can be derived by considering the TEM sample geometry as being either flat or wedge shaped. On the other hand, by considering a-priori knowledge about the sample chemistry measured intensities can be used to retrieve geometrical structures on the atomic scale. In particular, it is possible to derive the evolution of the APB with respect to one of the surfaces of a STEM specimen. This becomes clear when one considers the case of GaP on silicon where the constituents are known but the geometry of the crystal defect is to be investigated. Since the HAADF STEM signal is related to the atomic number *Z*, one can directly correlate the scattering strength to a chemical composition. This is in particular applicable in the case of GaP since the pure GaP mainphase (MP) is separated from the pure GaP antiphase (AP) only by their inner interface (APB). The compositional transition from the MP to the AP is illustrated in Fig. 1a, as cross-section along the [−110] direction. Due to the polar basis of GaP, single steps of the silicon substrate cause the creation of APBs. In contrast to the simplistic illustration in Fig. 1a the actual geometry of APBs is rather complex but can be considered to be monotonously changing from the MP to the AP as can be seen from previous experimental results (Beyer et al., 2012; Volz et al., 2011). Generally, a single crystal column seen in transmission can contain different fractions of MP and AP. Here, the boundary can be considered within every single column independently. Connecting these single column boundaries results in the overall appearance of the APB. This situation is illustrated in Fig. 1b where a cross-section along the [100] direction shows the chemical transition from MP to AP on a single column with an increasing fraction of the AP (gray shaded). On an atomic scale, this corresponds to a changing group-III and group-V sub-lattice decoration from gallium (Ga) to phosphorous (P) and from P to Ga respectively. Since this behavior is identical for any shape of the overall APB, the composition of each column can be used to map the z coordinate of the APB.

Seen along a low order zone axis like [001] as in the case of a plan-view (PV) investigation the atoms are aligned in columns along the z direction. The imaging plane of the microscope is therefore the xy plane.

2. Material and method

For the recording of high quality STEM measurements, several prerequisites were carried out in order to reduce the effects of beam damage and bending for a sufficiently large field of view. In the following, the preparation procedure will be explained and the STEM ADF measurements will be elucidated.