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Morphology and structure of thin epitaxial $Cu(In,Ga)S_2$ films on Si substrates

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Abstract

$Cu(In,Ga)S_2$ (CIGS) thin films were grown epitaxially on Si substrates of various orientations. The sulphur-termination process, utilized to suppress the unfavourable native surface reconstructions of Si, was examined. Thereby, an anisotropic etching of Si by sulphur was observed at high substrate temperatures. $Cu(In,Ga)S_2$ was found to show a strong tendency for nucleation on tips and at step edges of the substrates. Furthermore, the material was observed to agglomerate to existing grains as well. These mechanisms lead to a three-dimensional growth mode. The resulting roughness showed a strong and monotonic dependence on the Ga content of the samples, whereas the influence of the lattice mismatch on the morphology of the samples was found to be less distinct. The coexistence of the highly ordered chalcopyrite structure with the metastable CuAu structure was found to be another prominent feature in epitaxial $Cu(In,Ga)S_2$ thin films on Si.
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Keywords: Sulphur; $Cu(In,Ga)S_2$ thin film; Chalcopyrite structure

1. Introduction

Within the Cu-based family of chalcopyrite semiconductors, the sulphur compounds $CuInS_2$ (CIS) and $CuGaS_2$ (CGS) have attracted much attention in recent years. On one hand, they offer the perspective for cheap large area and completely nontoxic thin film photovoltaic devices. On the other hand, they can be grown epitaxially on silicon substrates [1–5]. Thus, $Cu(In,Ga)S_2$ (CIGS) bears the possibility for the monolithic integration of semiconductors with direct band-gaps into Si technology. However, it is well known that the performance of such heterojunction devices depends critically on the interface between epilayer and substrate as well as the morphology and structure of the grown films. Here, the quaternary system CIGS offers the possibility to grow lattice matched layers, in order to reduce the density of mismatch driven defects like dislocations to a minimum. Depending on the growth direction, perfect lattice match is expected for Ga fractions between 0.41 and 0.52 (for a-axis and c-axis orientation, respectively) [6]. Within this compositional range, preferably, CIGS thin layers with thicknesses between 1 nm and 1 μm have been grown on Si substrates with (111) and (100) orientation, respectively, in order to gain insight into the influence of the lattice mismatch on the morphology and structure of the samples. The initial stages of growth, the morphology, and structure of CIGS epilayers were examined by atomic force microscopy (AFM) and Rutherford backscattering spectroscopy (RBS). Additional information on the structure was obtained by in situ reflection high-energy electron diffraction (RHEED).

2. Experimental

All samples presented in the present study were grown by molecular beam epitaxy (MBE) from elemental sources. The metals (purities 99.9999%) were evaporated from commercial hot-lip effusion cells (Fa. *VTS CreaTec*). The sulphur (purity 99.9995%) was provided by a three-stage cracker source based on our own design [7]. The Si substrates were commercial, B-doped wafers of 100-mm diameter and either (111) orientation with 4° off-cut in <110>-direction or (100) orientation without off-cut. No ex situ chemical etching or rinsing was performed on the Si wafers; instead, they were cleaned and terminated in a

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high-temperature sulphurisation step described below. During film growth, the substrates were rotated and kept at temperatures of typically 830 K. The samples were examined ex situ with Rutherford backscattering spectroscopy (RBS) at the Jena University Laboratory for ion acceleration (*JULIA*) with an incident α-energy of 3.5 MeV. From the RBS-spectra, the composition, thickness, homogeneity, and surface roughness of the samples were deduced by a numerical evaluation of the spectra using the computer code RUMP [8,9]. Atomic force microscopy (AFM) measurements were performed using a commercial microscope (*Zeiss Veritek*). All AFM images in this work are depicted with a linear greyscale on the right side as a height scale. The RHEED measurements were done with 30 keV electron energy and a CCD-based data acquisition system (Fa. *Staib*).

3. Results and discussion

Prior to CIGS deposition, the Si surfaces were terminated in a high-temperature step with sulphur in order to clean the substrate and to suppress its unfavourable native surface reconstructions. This process yields Si(111)(4×4)-S and Si(100)(1×1)-S surfaces, respectively, which have turned out to be highly suitable for the subsequent epitaxial growth of CIGS thin films [1,3]. As an enhancement to our previous works, the sulphur termination is now monitored by a residual gas analyser (RGA). Fig. 1 depicts the temperature profile for the sulphur termination of a Si(111) substrate along with the total pressure and the partial pressure at 60 amu as measured by a vacuum gauge and the RGA, respectively. A drastic increase of the RGA signal at substrate temperatures above 1173 K is attributed to the onset of an effective etching of the Si surface by sulphur and the subsequent desorption of SiS. This etching holds on at lower substrate temperatures, although the desorption rate decreases with decreasing substrate temperature. It takes

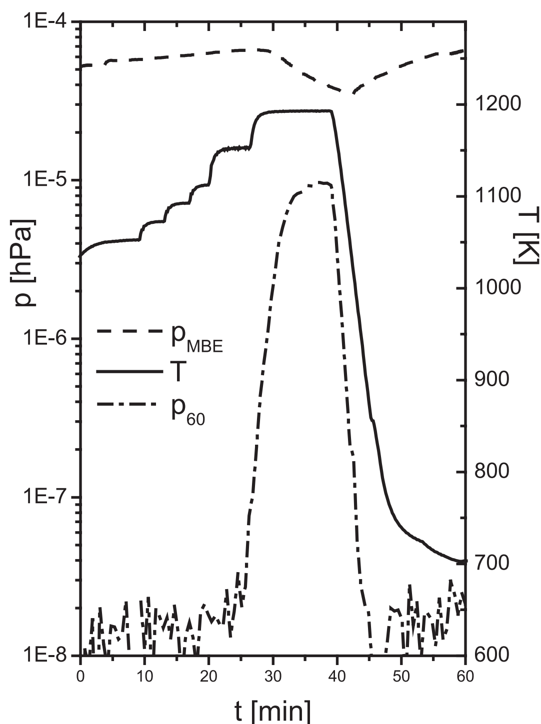

Fig. 1. Temperature profile and pressures during the S-termination of a Si(111) surface. The onset of the partial pressure of the mass 60 amu is attributed to the formation of volatile SiS.

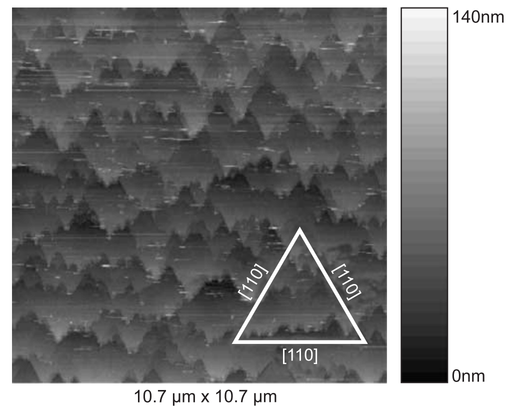

Fig. 2. AFM scan of a Si(111) surface after etching with a linear greyscale, representing the height of the surface steps. The triangular incisions are due to an anisotropic etching by elemental sulphur at high temperatures.

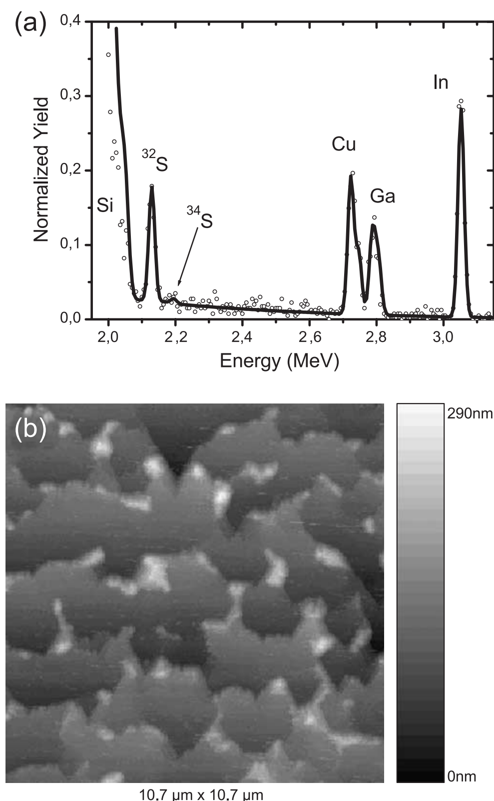

Fig. 3. RBS spectra (a) and AFM image (b) of a CIGS thin film on Si(111). The evaluation of the RBS spectrum yields a homogeneous film thickness of 2.5 nm and a Cu-rich composition ($Cu_{1.17}In_{0.41}Ga_{0.42}S_{2.1}$). Note that the natural isotope distribution of the elements can easily be detected by the splitting of the S-yield, the step visible in the Cu-yield, and a slight asymmetry of the Ga-yield. The AFM picture (b) reveals that the epilayer does not cover the whole surface, but instead tends to agglomerate at step edges and tips of the substrate.

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