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ScienceDirect
Journal of Non-Crystalline Solids 354 (2008) 2310–2313
JOURNAL OF NON-CRYSTALLINE SOLIDS
www.elsevier.com/locate/jnoncrysol

Microscopic study of the H_2O vapor treatment of the silicon grain boundaries

S. Honda *, T. Mates, B. Rezek, A. Fejfar, J. Kočka

Institute of Physics, Academy of Sciences of the Czech Republic, Cukrovarnická 10, 162 53 Prague 6, Czech Republic

Available online 20 February 2008

Abstract

We have proposed annealing in H_2O vapor as a new effective and low-cost method for passivating polycrystalline silicon grain boundaries and for improving the performance of poly-Si based devices. The effect of H_2O vapor treatment was experimentally found to differ from analogous anneal in nitrogen and hydrogen. Mechanism of the H_2O vapor treatment was studied by Kelvin force microscopy, used to measure the potential change at individual grain boundaries and point defects. The potential change was dependent on the grain boundary character and it correlated with the crystalline disorder and internal stress observed by microscopic Raman spectroscopy. Effect of H_2O vapor passivation was experimentally connected with the potential change at the grain boundaries before and after the treatment.
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PACS: 73.50.−h; 73.40.−c; 67.80.Mg; 73.40.Cg; 73.40.Mg; 81.40.−z

Keywords: Polycrystalline silicon films; H_2O vapor treatment; Potential; Crystalline disorder; Stress; Defects; Passivation; Micro Raman spectroscopy; Kelvin force microscopy

1. Introduction

Performance of polycrystalline silicon devices largely depends on the transport properties limited by the grain boundaries with associated potential barriers and defect states. In order to decrease the influence of grain boundaries it is necessary to passivate them, usually by plasma hydrogenation. We have recently proposed H_2O vapor treatment as an alternative passivation method [1]. Treatment by H_2O vapor offers advantages for solar cells industry, being a very simple, non-toxic process with a good uniformity even in large volumes.

Although improvement of the Si electronic properties by the H_2O vapor treatment was already demonstrated [1,2], the underlying mechanism remains unclear. Electronic activity associated with the Si grain boundaries depends on the grain boundary character, defects and impurities, stress and other factors, complicating the search for H_2O treatment mechanism. Therefore a microscopic characterization of the grain boundaries and their electronic properties is required. Electronic activity of the individual Si grain boundaries has been studied for a long time using electron beam induced current, revealing several kinds of grain boundaries (Σ3, Σ9 and random) with different recombination velocities [3]. Recently it was found that grain boundaries do not always act as effective recombination site for carriers [3].

Here we tried to correlate the potential change and crystalline disorder and/or stress at the grain boundaries using Kelvin force microscopy (KFM) and microscopic Raman spectroscopy. The results are discussed as the first experimentally based explanation of the H_2O vapor treatment mechanism.

2. Experiment

Poly-Si thin films with ~15 μm thicknesses were grown by atmospheric pressure chemical vapor deposition (APCVD) with SiH_2Cl_2 source gas and BCl_3 as doping

* Corresponding author. Tel.: +420 220 318 519.
E-mail address: honda@fzu.cz (S. Honda).

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doi:10.1016/j.jnoncrysol.2007.09.107

gas directly on thermally oxidized Si wafer substrates kept at 1050 °C [4]. Average grain size was around 2–3 μm. Doping concentration of the film was 10^{17} cm^{-3}. For comparison we have used polished p-type bulk multicrystalline Si. Defect etching was performed by Secco etching ($K_2Cr_2O_7$ (0.15 mol/l):HF (50%) = 1:2) for 5 min at room temperature [5]. The electronic properties of the poly-Si thin films were characterized by Hall effect in van der Pauw configuration with contacts formed after removing any oxide resulting from the treatment.

Nitrogen, hydrogen and H_2O vapor treatments were performed at atmospheric pressure or at high pressure (0.7 MPa). In the case of high pressure treatment we placed the samples in a chamber filled with H_2O vapor and heated in a furnace, using the amount of water to determine the pressure inside the tube. Treatment temperature was varied from 150 °C to 900 °C. All samples were treated for a fixed time of 1 h.

Raman spectra of multi-Si wafers were measured at ambient conditions using Renishaw inVia Reflex Raman microscope in a backscattering geometry with 785 nm laser excitation. Kelvin force microscopy (KFM) was measured at room temperature and ambient conditions by Veeco Dimension 3100 atomic force microscope (AFM) using PtIr coated cantilevers (75 kHz).

3. Results

3.1. H_2O vapor treatment for poly-Si thin films

Fig. 1 shows the Hall mobility after treatment by different atmospheres (H_2O vapor, D_2O vapor, hydrogen and nitrogen) at 300 °C as a function of treatment time, as reported by us at the last conference [1]. The remarkable improvement of the Hall mobility was repeatedly confirmed by the following experiments aimed at comparing the effect of H_2O vapor and other atmospheres.

Fig. 2 shows the Hall mobility after nitrogen anneal and subsequent H_2O vapor treatment. Temperature was varied

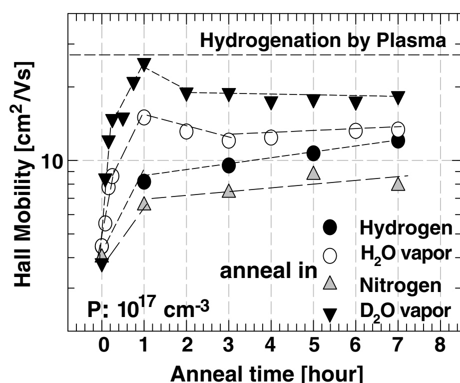

Fig. 1. Hall mobility after anneal at different atmospheres (at 300 °C and atmospheric pressure) as a function of the annealing time. The values can be compared to the mobility reached after optimized 1 h plasma hydrogenation marked by dashed horizontal line [1].

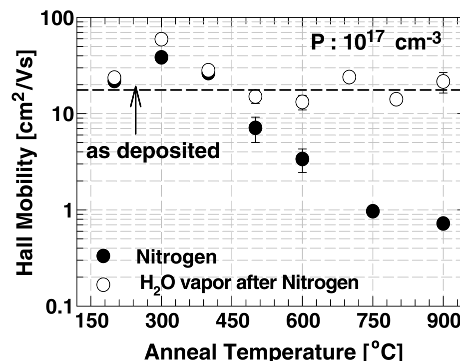

Fig. 2. Hall mobility after anneal in nitrogen and after subsequent H_2O vapor treatment.

from 200 °C to 900 °C during the 1 h nitrogen anneal. After nitrogen anneal, the same samples were treated for 1 h by H_2O vapor at 300 °C at atmospheric pressure. The Hall mobility increased with an increase of the nitrogen temperature upto 300 °C. However, with further temperature increase the mobility started to decrease down to 0.7 cm^2/Vs. The poly-Si thin films annealed in nitrogen were then treated by H_2O vapor. The Hall mobility decreased by nitrogen anneal at temperatures >500 °C recovered up to the value of the as-deposited film. The same recovery was observed when plasma hydrogenation was used after the nitrogen anneal.

H_2O vapor treatment was also compared with the hydrogen anneal at temperatures from 150 °C to 550 °C. After hydrogen anneal, subsequent H_2O vapor treatment was performed on poly-Si thin films at the same temperature in atmospheric pressure. Fig. 3 shows the Hall mobility after hydrogen anneal and also after subsequent H_2O vapor treatment as a function of the temperature.

Hall mobility first increased with the hydrogen anneal temperature and then started to decrease above 350 °C. After the subsequent H_2O vapor treatment, Hall mobility increased at all annealing temperatures, reaching the highest value of 57 cm^2/Vs at 450 °C. This indicated that the H_2O vapor treatment still could improve Hall mobility after hydrogen anneal.

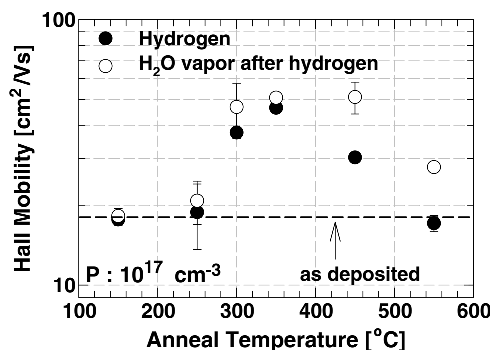

Fig. 3. Hall mobility after hydrogen and after subsequent H_2O vapor treatment.

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