Detection of Hydrogen on Grain Boundaries of CVD-Grown Diamond

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Diamond with its hardness and outstanding electronic, optical and chemical properties is a very interesting material for industry and research. Chemical vapour deposition (CVD) is a technique to produce hetero-epitaxial grown polycrystalline diamond layers. These layers contain small amounts of hydrogen in the ppm-range which has critical influence to electronic and optical properties. The content decreases with larger crystallites which indicates that a large part of the hydrogen is situated at the grain boundaries. However, with available hydrogen analysis methods no direct measurement could be performed to prove this speculation and determine the amount of hydrogen sitting on grain boundaries.

Now, for the first time, we were able to detect hydrogen localized at grain boundaries inside a near $< 110 >$-textured layer. This sensitive analysis of hydrogen with a spatial resolution better than 1 μm necessary for this measurement becomes possible due to the new nuclear microprobe SNAKE (Superconducting Nuclear nanoprobe for Applied nuclear (Kern-) physics Experiments) at the Munich tandem accelerator [1,2,3]. For this purpose, we applied the technique of coincident elastic proton-proton scattering [4] using a sub-micrometer focused 17 MeV proton beam. [6,5] An annular silicon strip detector covering a solid angle of detection of 2.3 sr is used for coincidence and energy analysis. [7] The analysis filters coincident protons in opposite of 16 sectors in a window of about 60 ns. Additionally, 48 rings with a 10 mrad angular resolution enable a second filter looking for an angle of 90° between the scattered protons. Using the energy information, the correlation between energy difference and scattering angle of coincident protons is used for the third filter stage. Finally, an advanced time analysis of the sector signals of the coincident protons by TDCs with a time resolution of about 3 ns is applied as a fourth filter.

With these four filters, it is possible to suppress accidental coincidences by a factor of 10^{-7} even at total count rates of 100 kHz. Therefore, we were able to detect hydrogen with micrometer resolution and ppm sensitivity in a 55 μm thick diamond layer. In the optical microscope image of Fig. 1b the marked area shows a line scan of 7 $\mu m \times 70$ μm across the visible grain boundaries. In Fig. 1a the detected hydrogen content inside the layer results in discrete peaks at the positions of the optical structures. This hydrogen enhancement is even more obvious in Fig. 1c with the coincidence events plotted in dependence of depth and position of the layer. 1-2 monolayers of hydrogen contamination at the surfaces results in the two horizontal branches. Between this surfaces the hydrogen is located at the grain boundaries one perpendicular and the other tilted towards the surface. A hydrogen content of one tenth of a monolayer or 3×10^{14} at/cm^2 projected to the face of the grain boundaries is obtained. Inside the grains the hydrogen content was found to be below the detection limit of 0.1 at-ppm.

This successful detection of hydrogen on grain boundaries of CVD-diamond demonstrates the unique potential of this three dimensional hydrogen microscopy at SNAKE.

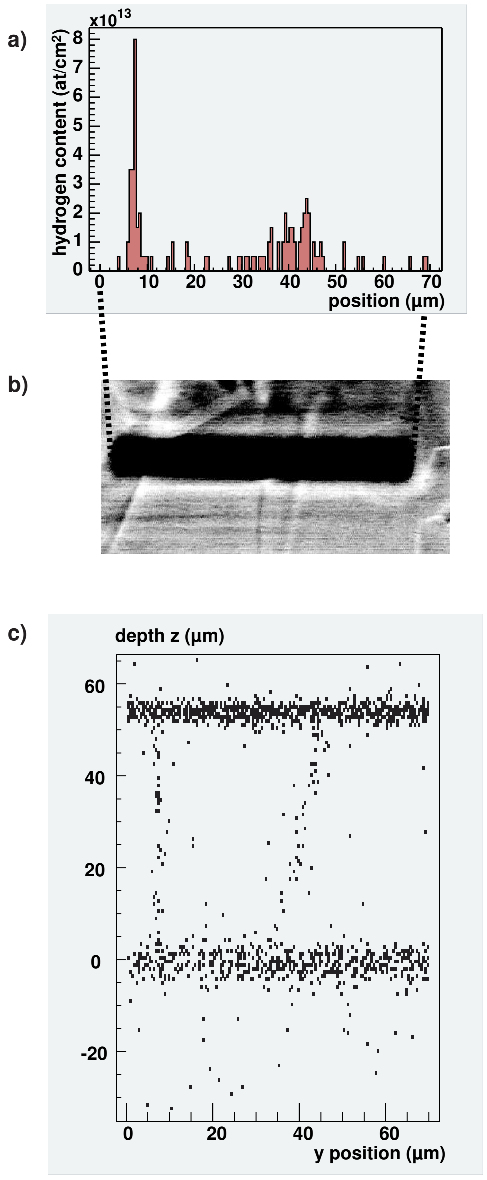

Fig. 1: a) Hydrogen content inside the layer depending on the position of the 7 $\mu m\times70$ μm line scan shown in the optical microscope image b). A large enhancement of hydrogen at the position of the left structure is clearly visible. The broader right peak also matches with the position of the right two lines in the optical picture. The coincident proton events plotted in a depth profile c) of the line scan shows the hydrogen enhancement at the grain boundaries inside the layer: the horizontal braches raise due to 1-2 monolayers of hydrogen contamination at the front and rear suface. Between this surfaces the hydrogen is located at the grain boundaries one perpendicular and the other tilted towards the surface.

References

[1] G. Datzmann, G. Dollinger, Annual report 2001, p. 62
[2] G. Datzmann *et al.*, Annual report 2001, p. 63
[3] G. Datzmann, Ph.D. thesis, TU München, 2002
[4] B. L. Cohen *et al.*, J. Appl. Phys. **43** (1972) 19
[5] G. Dollinger *et al.*, Apl. Phys. Lett. **82(1)** (2003) 148
[6] P. Reichart *et al.*, Nucl. Instrum. Methods B **197** (2002) 134
[7] P. Reichart *et al.*, Annual report 2001, p. 66