TEM investigations of ZnO nanocrystals embedded in SiO_2

R. Schneider[1], G. Mayer[2], M Fonin[2], U. Rüdiger[2], N. Janßen[3], R. Bratschitsch[3], and D. Gerthsen[1]

1. Laboratory for Electron Microscopy, University Karlsruhe, D-76128 Karlsruhe, Germany
2. Department of Physics, University Konstanz, D-78457 Konstanz, Germany
3. Department of Physics, University Konstanz and Center for Applied Photonics, D-78457 Konstanz, Germany

reinhard.schneider@kit.edu
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Quantum dots have attracted considerable attention because the spins of excess electrons in single-electron quantum dots are potential candidates for qubits in quantum information technology (cf., e.g., [1]). In our study, ZnO nanocrystals (NCs) were prepared via radio-frequency magnetron sputtering as a SiO_2/ZnO/SiO_2 trilayer on an Si(100) substrate with an intermediate in-situ annealing step (for more details see [2]).

In order to reveal the structural properties of the ZnO/SiO_2 composite system, detailed TEM investigations were performed using a Philips CM 200 FEG/ST at 200 kV accelerating voltage. Fig. 1a shows a representative cross-section TEM image of the approximately 5 nm thick ZnO layer embedded in SiO_2. From this image the presence of slightly elongated ZnO NCs (dark regions) dispersed within the amorphous SiO_2 surrounding (bright background) can be assumed. In Fig. 1b an HRTEM image of the ZnO layer is shown, confirming its crystalline quality. To verify the formation of separated ZnO NCs, energy-filtered TEM and high-angle anular dark-field (HAADF) STEM were carried out on plan-view samples using a 200 kV LEO 922 Omega. A typical bright-field TEM image is shown in Fig. 2a, suggesting the presence of individual NCs. The corresponding element map (Fig. 2b) obtained with the Zn-$L_{2,3}$ ionization edge (threshold energy of 1020 eV) using the three-window technique clearly visualizes a strongly inhomogeneous Zn distribution, indicating distinct ZnO NCs surrounded by SiO_2. In Fig. 2b the Zn-enriched regions have an extension of about 15-20 nm. However, because of the low signal-to-noise ratio of the Zn map the size of the ZnO regions can be broadened. HAADF STEM imaging (Fig. 2c) with its high atomic-number contrast exhibits bright regions that can be correlated to ZnO crystallites which are separated by intermediate darker regions of SiO_2. In addition, about 1000 NCs from several plan-view TEM images were analyzed to obtain the size distribution of the ZnO NCs, yielding that 73.4% of the NCs are 5 ± 2 nm in diameter. Only 3% of the NCs are smaller than 3 nm, 24.6% of the NCs are larger than 7 nm.

Fig. 3a shows a Debye electron diffraction pattern of the ZnO/SiO_2 system taken from a plan-view sample, where the corresponding radial intensity profile after background subtraction (Fig. 3b) is given together with a simulated powder diffraction diagram of the ZnO wurtzite structure. Applying the Scherrer formula $D = 0.9\,\lambda$ / (FWHM cos θ) that is well known from X-ray diffraction [3] with FWHM representing the full width at half maximum (in radians) of the investigated diffraction peak the mean grain size D of the NCs may be calculated. In this formula λ denotes the electron wavelength and θ the Bragg angle. Evaluating the strong and well separated 110, 103, and 112 reflections a grain size of 5 ± 1 nm was found, which is in good agreement with the above-mentioned size distribution. Plan-view HRTEM imaging confirms the presence of crystalline, predominantly small ZnO NCs with sizes up to 16 nm, embedded in amorphous SiO_2. Fig. 3c shows an HRTEM image of a representative ZnO NC with a diameter of about 10 nm and the expected wurtzite

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structure. High-resolution TEM imaging on different NCs does not reveal a preferential alignment of the ZnO NCs along the growth direction.

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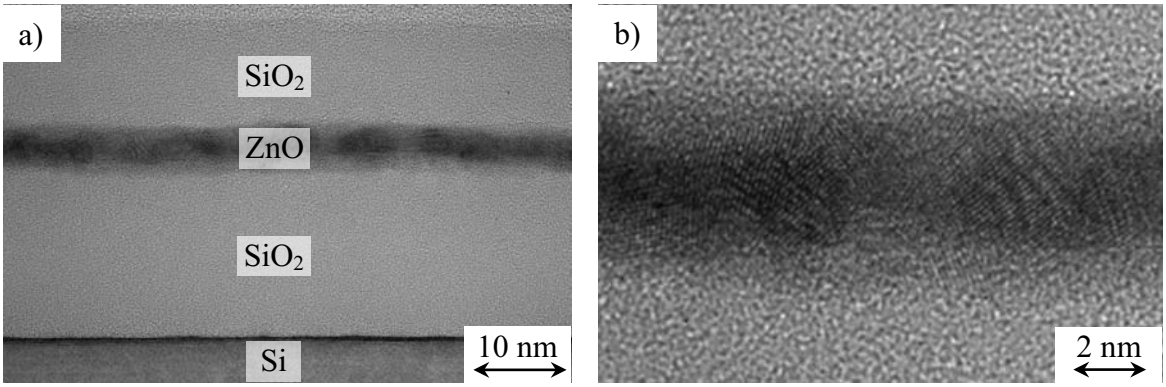

Figure 1. a) Cross-section TEM image of the ZnO layer embedded in SiO_2, b) detailed HRTEM image of the ZnO layer.

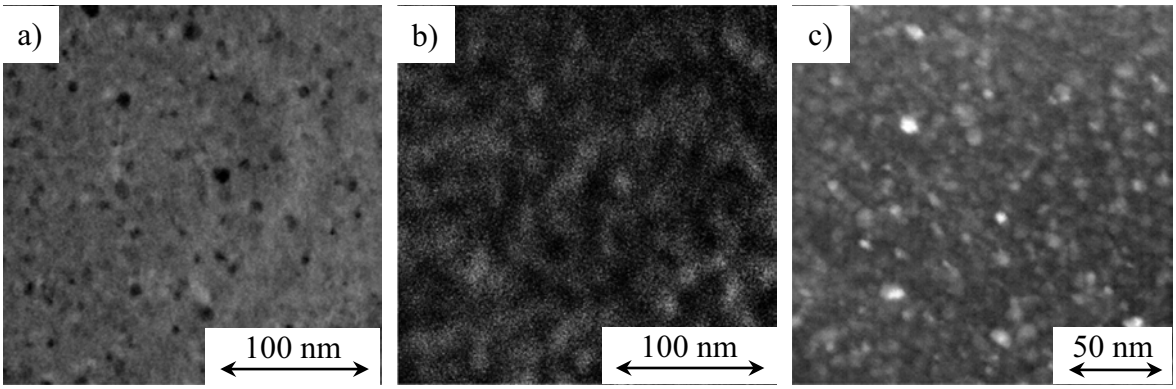

Figure 2. a) Plan-view bright-field TEM image of the ZnO/SiO_2 system, b) corresponding EFTEM image of the Zn distribution, and c) HAADF STEM image.

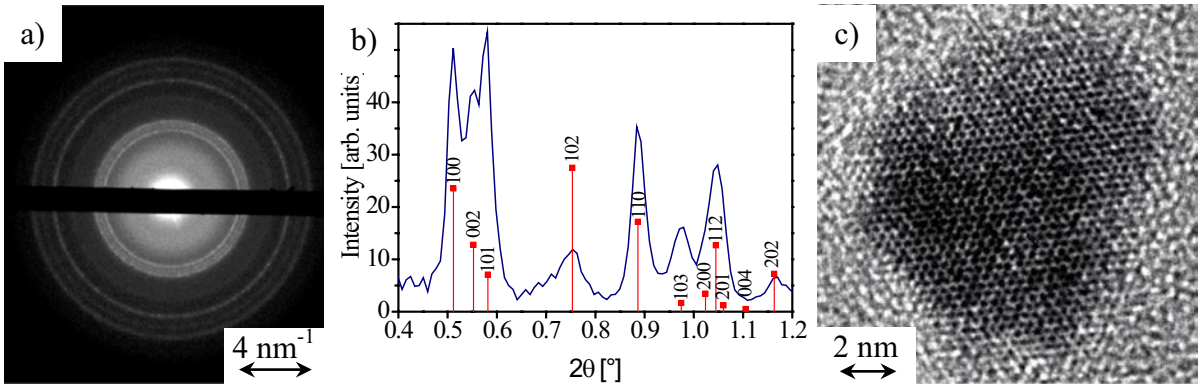

Figure 3. a) Electron-diffraction pattern of the ZnO/SiO_2 composite system, b) comparison of the radial intensity profile of (a) (solid line) with a simulated powder diffraction diagram (indexed bars), and c) plan-view HRTEM image of a single ZnO NC.

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