STEM-based direct observation of dislocation-pipe diffusion in metal/semiconductor nitride superlattice thin films

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Device failure from diffusion short circuits in microelectronic components occurs via thermally induced migration of atoms along high-diffusivity paths: dislocations, grain boundaries, and free surfaces [1-3]. Knowledge about the structural features along which diffusion paths in solids are formed is hence of great importance. For instance, kinetic processes limited by grain-boundary and/or dislocation diffusion in materials are known to be diffusion creep, precipitation, coarsening, solute segregation, strain aging, grain boundary migration, and sintering. Even well-annealed single-grain metallic films contain dislocation densities of about 10^{14} m^{-2}; hence dislocation-pipe diffusion (DPD) becomes a major contribution at working temperatures. While its theoretical concept was established already in the 1950s [4] and its contribution is commonly measured using indirect tracer, spectroscopy, or electrical methods [5], no direct observation of DPD at the atomic level has been reported.

Aberration-corrected STEM with an image- and probe-corrected and monochromated FEI Titan[3] 60-300 kV instrument equipped with a high-brightness XFEG source and Super-X EDS detector for ultra-high count rates, operated at 300 kV, was employed [6]. We present atomically-resolved STEM images of the onset and progression of diffusion along threading dislocations in sequentially annealed nitride metal/semiconductor superlattices, which are a class of materials of high interest due to their high hardness and thermal stability, as well as plasmonic properties [7-13]. The STEM micrographs are accompanied by EDS maps and GPA analysis (Fig 1). From the images showing the same region at different time-steps during annealing, diffusivity coefficients are calculated directly. Most importantly, we show that this type of diffusion is independent of concentration gradients in the system but governed by the reduction of strain fields in the lattice [14]. Our images of the formation of pipes around the dislocation core and at different time steps in an annealing series are the first direct experimental observations of this phenomenon at atomic resolution employing TEM techniques.

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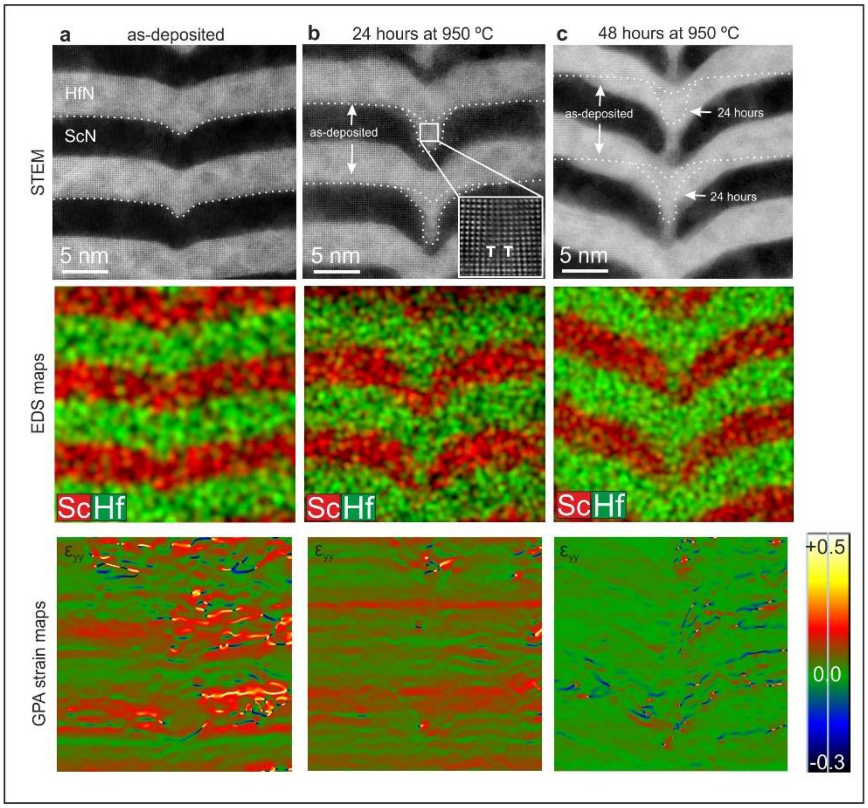

Fig. 1. The operation of dislocation-pipe diffusion by strain fields around a threading dislocation line. High-resolution STEM micrographs, corresponding EDS- and strain mapping of the same area of a HfN/ScN superlattice sample as-deposited (a), and after annealing for 24 h and 48 h, respectively, at 950 °C (b) and (c). The onset of Hf diffusion along the dislocation line after deposition is already visible in a. From the change in the shape of the Hf diffusion front after annealing in b, the diffusion length can be directly measured and an average value calculated. The enlarged region in the inset in (b) shows pairs of edge dislocations at the cores of the vertical dislocation line, in the center along which the diffusion occurs. Strain mapping reveals high strain fields around the dislocation line of the as-deposited sample a, that become significantly reduced by the diffusion of Hf after 24 h of annealing (b), and relaxes the lattice almost entirely once the pipe formation is completed (c). Shown is the y-component of the strain tensor ε_{yy}, i.e. parallel to the direction of diffusion.