Carbon at Si (111)-twins: TEM analysis supported by molecular dynamics structure relaxations

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High carbon supersaturation and high density of twin lamellae in multi-crystalline silicon (mc-Si) may influence material properties and applicability in photovoltaics [1]. To understand e.g. structure modifications and stress relief total energy minimizations by molecular dynamics (MD) simulations at ab initio level or using empirical potentials have been employed to describe the carbon incorporation in silicon. Through a combination of TEM, HREM (JOEL JEM 4010), EDX (Phillips CM 20 FEG), and image contrast interpretations based on the MD-relaxed structure models the accumulation of carbon at (111)-twin boundaries can be analyzed [1,2].

For small silicon (111)-twin structure models (cf. Figure 1) quantum-theoretical ab initio calculations are applied (CASTEP-DFT, LDA or GGA, norm-conserving pseudopotentials, 280eV energy cutoff). Starting from a geometric twin model (24 atoms in a 6.65Åx18.81Åx3.84Å supercell, all bond lenghts 2.35Å) the C atoms are substituted at one of the twin planes (b,f) or both layers (e) are replaced. For comparison C is also substituted in layers parallel to the twin (c) or randomly in the bulk (d,f). Different processes (minimization or dynamics) and boundary conditions (various fixed vs. variable supercell borders) yield to different structure reorientations (Figure 1 shows typical resulting bond lengths and thus lattice fringe modifications at the twins). The energy gain for the models shown in Figures 1a) to f) is 0.34, 2.3, 1.6, 2.9, 6.1 and 5.8 eV/atom, respectively, compared always with the non-relaxed, geometric twin start model (however, d-f with all cell borders free).

For larger systems, as e.g. shown in Figure 2 with 54000 atoms and various twin layers, the only possibility of simulating time dependent atomic processes with macroscopic relevance is the molecular dynamics (MD) method solving Newton's equations of motion for all particles and using suitably fitted many-body empirical potentials, preferably of the Tersoff (TS) or bond-order type [3,4]. The calculations are done using a constant pressure (NpT ensemble, relaxation at 0K, and annealing up to 900K) and time steps of the order of 0.8 fs.

Figure 3 shows simulated 400kV-<110>-HREM images for a sample thickness t=11.5nm and two different defoci Δf = 0nm (i), 40nm (ii). The lattice relaxation can be found in the image contrast, but varying with the imaging conditions. Unique interpretations need a matching of thickness-defocus series. In addition, image simulations as given in Figure 5 may allow an explanation of the contrast variations along the experimental HREM micrograph in Figure 4, which occur probably due to C precipitation, remaining strains or bended samples. The 400kV HREM micrographs of Figure 5 are simulated with the same parameters as of Figure 3, but with the structure model of Figure 2 after annealing up to 900K. The contrast variations are here yielding from carbon rearrangements at the interfaces and resulting deformations.

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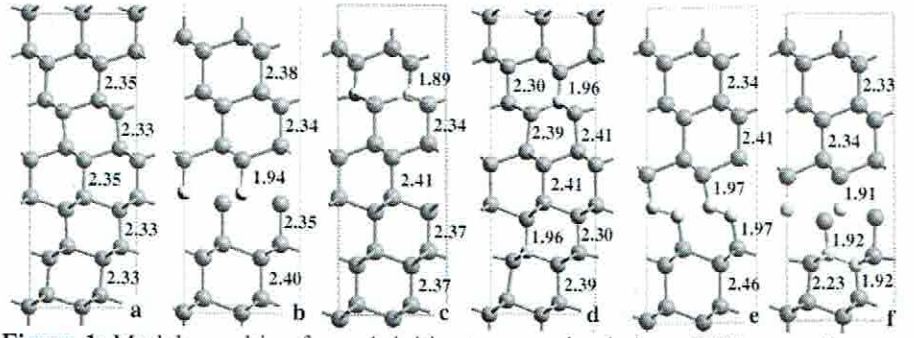

Figure 1: Models resulting from ab-initio structure simulations (DFT approximation, cf. text) to minimize the total energy: a) Si twin structure with a {111} lattice plane distance of 3.14Å (numbers indicate the important next neighbor distances in Å); b) Half of (111) twin double plane occupied with C (brighter spheres); c) A C-layer parallel to the twin; d) Randomly distributed C substitutions; e) Double layer of C occupation at the twin yielding an additional lattice shift; f) Half twin C occupation and randomly distributed C near the twin

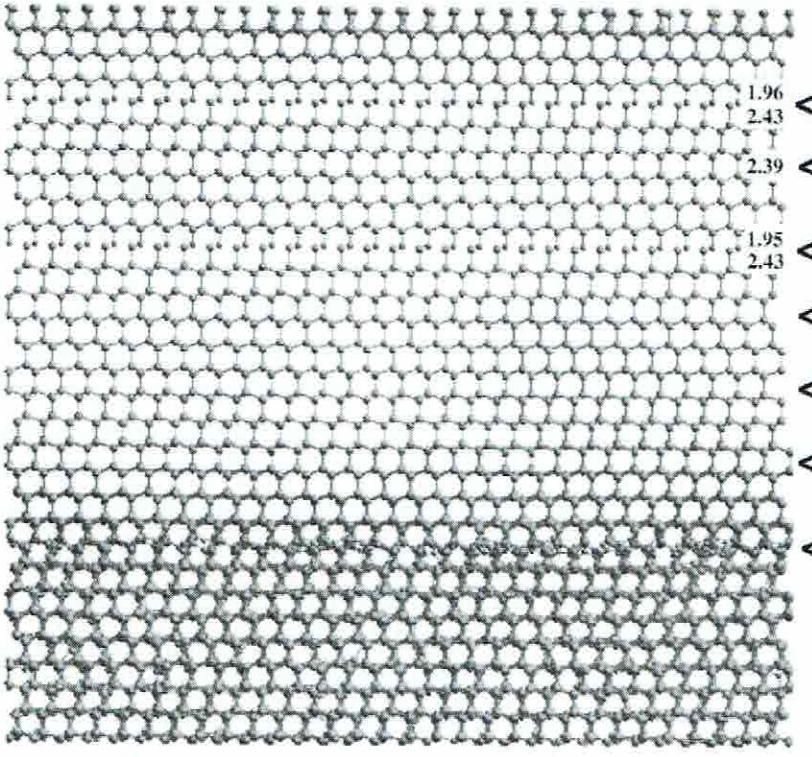

Figure 2: MD-relaxed model (54000 atoms, TS potential) with (111)-twins (T), half (C) or double (2) layer C-occupation, and random C-substitution (R).

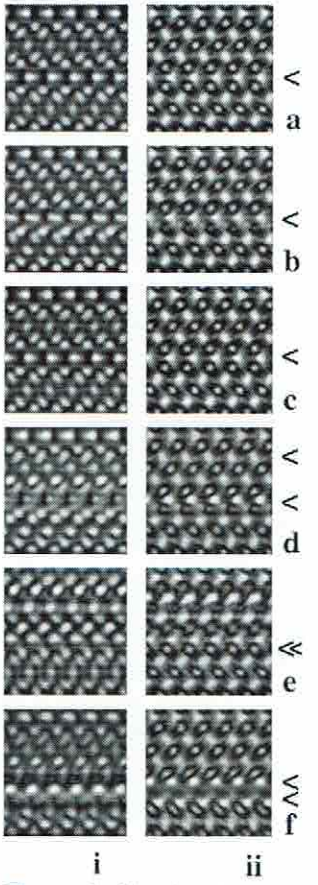

Figure 3: Simulated 400kV-<110>-HREM images of the relaxed twins corresponding to the models of Fig. 1 with differently incorporated C atoms (t=11.5nm, Cs=1mm, defocus spread 10nm, beam divergence 0.5mrad, i: Gauss focus, ii: Scherzer Focus; twins or C marked by arrows).

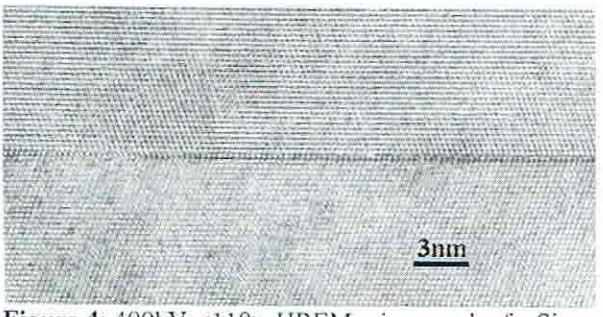

Figure 4: 400kV-<110>-HREM micrograph of a Si-{111}- twin with varying contrast due to bending of the sample, carbon segregation and/or strains.

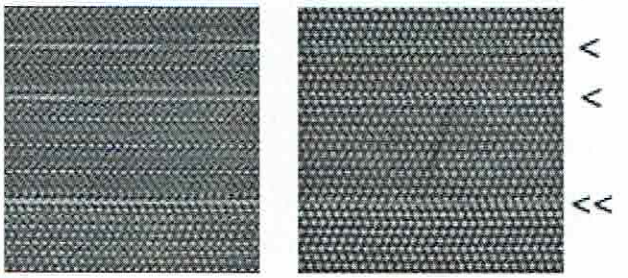

Figure 5 Simulated HREM of <110> oriented twins in the relaxed structure of Fig. 2 (C indicated; left: Gauss-right: Scherzer-focus, parameter as Fig. 3).