Ultrafast optical excitation of a metastable, long-living, and super cooled state in Peierls distorted Indium atomic wires on Si(111)

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We employ femtosecond-reflection high energy electron diffraction at an ultimate temporal resolution of 330 fs [1] to follow the structural dynamics during the optically driven 1st-order phase transition in the prototypical indium atomic wire system on a Si(111) surface. The lifting of the Peierls distortion and transition to the high symmetry (4×1) state occurs by an accelerated displacive excitation scenario in less than 700 fs [2]. The immediate recovery to the (8×2) ground state is hindered by a significant energy barrier along the reaction coordinate from the excited (4×1) state to the (8×2) ground state [3]. Any excess energy originating from the optical excitation through a 800 nm 80 fs laser pulse is extremely efficiently dissipated into the Si substrate [4]. As a result the excited metallic (4×1) high temperature state is trapped in a supercooled metastable phase for many nanoseconds. The recovery to the insulating (8×2) ground state is facilitated through nucleation at individual atomic steps – acting as seeds for the ground state - on the slightly miscut Si surface. From an average terrace width of 300 nm together with the recovery time constant of 3 ns we obtain a speed of the recovery front – which propagates 1-dimensionally like a row of falling dominoes – of v_{rec} = 112 m/s [3,5]. The influence of the degree of super cooling on the speed of recovery front will be discussed.

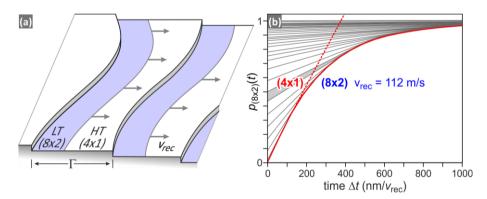


Fig.1: (a) scetch of slightly miscut and atomically stepped Si surface during recovery of the (8×1) groundstate subsequent to the excitation of the (4×1) state. Steps separating terraces of width Γ act as seeds for 1D recovery fronts travelling at a speed v_{rec}. (b) The terrace width distribution p(Γ) enters into the evaluation of the speed v_{rec} of recovery front.

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