Supporting Information: Dynamic Catalyst Restructuring During Carbon Nanotube Growth

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Figure SI1 shows evidence for lattice fringes in the Ni clusters at temperatures as high as$\sim$890K. Other examples can be found in Ref. 1.

Effect of dissolved C

The possible presence of dissolved C does not affect the conclusions of our continuum model. To verify this, we compute the diffusion coefficient of a fcc-Fe(110) surface with and without dissolved C. We use Fe instead of Ni due to the availability of tested Fe-C potentials in the literature, where Fe-Fe, Fe-C and C-C potentials are second moment tight-binding, Johnson and Lennard-

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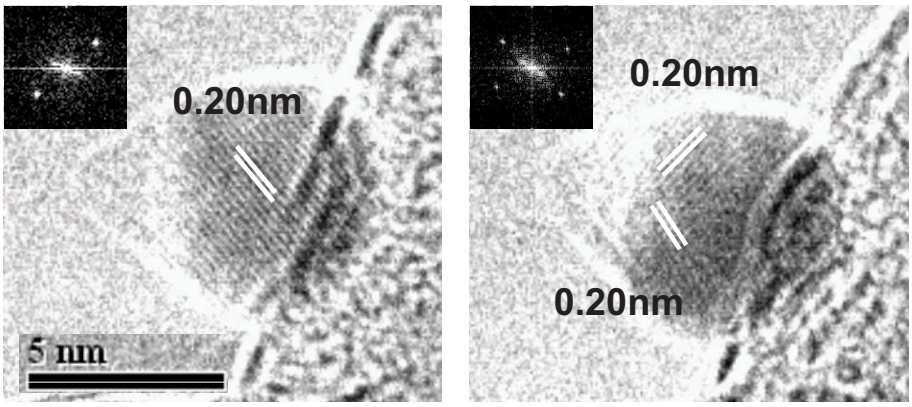

Fig SI1. ETEM snapshots of a Ni nanocrystal at$\sim$890K in $\sim 10^{-3}$mbar C_2H_2. The strong reflections in the images fast Fourier transforms (insets) correspond to fcc Ni$\{111\}$ planes.

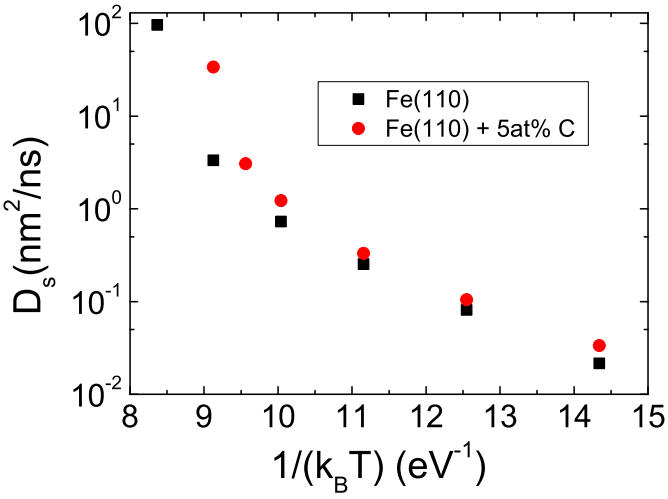

Fig SI2. Arrhenius plot showing the negligible effect of C for self-surface diffusion in a fcc-Fe(110) slab, periodic in "x" and "z". A wide vacuum layer avoids self interaction in the "y" direction. The diffusion coefficient is calculated with the drift force method.[3] The pristine slab consists of 12 layers, 7 free to move. A force f=0.25eV/nm is applied to all Fe atoms in the upper free-layers in the "+z" direction. As a result, these acquire a velocity v. The surface diffusion coefficient is calculated as $D_s = vk_BT/f$ and plotted in black squares. When $\sim$5at% C is dissolved, the force on the Fe atoms is the same, while that on the C atoms is in the opposite direction to simulate the opposite flow of Fe and C during CNT growth and particle dewetting. D_s of the Fe-C system is represented by red circles.

Jones.[2] The behavior of C in Ni should follow the same trends as in fcc-Fe. Figure SI2 shows that dissolving up to $\sim$5at% C, the diffusion coefficient increases negligibly with respect to pure Fe.

Movies

Experimental

- **Movie S1**. Real time ETEM movie in 3:1 NH_3:C_2H_2 at 1.3mbar and 750K showing morphing of an elongated Ni particle dewetting from a CNF, with a final spherical shape. Fig. 1a in the main text plots the frames of this movie.

- **Movie S2**. Real time ETEM movie in 3:1 NH_3:C_2H_2 at 1.3mbar and 750K showing the escape of a Ni particle trough a carbon nano-neck. Fig. 1b in the main text plots the frames of this movie.

- **Movie S3**. Real time ETEM movie in 3:1 NH_3:C_2H_2 at 1.3mbar and 750K showing the splitting of a Ni particle due the formation of a carbon neck. Fig. 1c in the main text plots the frames of this movie.

Molecular Dynamics

- **Movie S4**. MD simulation of a Ni nanoparticle dewetting from a CNT via surface diffusion. Fig. 6a in the main text plots frames of this movie.

- **Movie S5**. MD simulation of a Ni nanoparticle dewetting from a CNT with a neck constriction. Fig. 6b in the main text plots frames of this movie.

- **Movie S6**. MD simulation showing the fracture of a back-anchored Ni nanoparticle caused by a Nichols-Mullins instability initiated by a neck constriction in the CNT. Fig. 6c in the main text plots frames of this movie.

References

1. Hofmann, S.; Sharma, R.; Ducati, C.; Du, G.; Mattevi, C.; Cepek, C.; Cantoro, M.; Pisana, S.; Parvez, A.; Cervantes-Sodi, F.; Ferrari, A. C.; Dunin-Borkowski, R.; Lizzit, S.; Petaccia,

L.; Goldoni, A. Robertson, J. Ledge-Flow-Controlled Catalyst Interface Dynamics During Si Nanowire Growth *Nano Lett.* **2007**, *7*, 602-608.

2. Ding, F.; Bolton, K.; Rosen, A. Nucleation and Growth of Single-Walled Carbon Nanotubes: A Molecular Dynamics Study. *J. Phys. Chem. B* **2004**, *108*, 17369-17377.

3. Mehrer, H. *Diffusion in Solids. Fundamentals, Methods, Materials, Diffusion-Controlled Processes*. Springer: Berlin, 2007.