WILDE LAB.

[Hydrogen Transport Mechanisms at Surfaces]

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Nanoscale Surface Physical Chemistry

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Absorption, Exchange, and Diffusion at $H_{(2)}$-Exposed Surfaces

Atomic Scale Clarification of Hydrogen Penetration, Exchange, Diffusion, and Catalytic Reaction Processes

Hydrogen (H) absorption and diffusion in and desorption from metals and oxide nanoparticles and thin films are crucially important for the storage and purification of H_2 in clean energy technology (fuel cells, fusion) and for industrial hydrogenation catalysis. Our research clarifies the microscopic pathways along which gas phase H_2 dissociates at the surfaces and H atoms penetrate into the interior of metals and oxides, and why Pd-absorbed H is essential for olefin (C=C) hydrogenation catalysis on Pd. To aid the development of novel efficient hydrogenation catalysts and hydrogen storage materials, we investigate through isotope labeling and H/D exchange experiments at pure and modified palladium (Pd) surfaces how the H transport across the gas/solid interface depends on the surface structure and thereby becomes controllable at the atomic level. We also study H diffusion in (photo)catalytic oxide thin films and the fusion-related H isotope (HI) retention in HI plasma-exposed tungsten.

◆ Experimental Techniques & Key Information

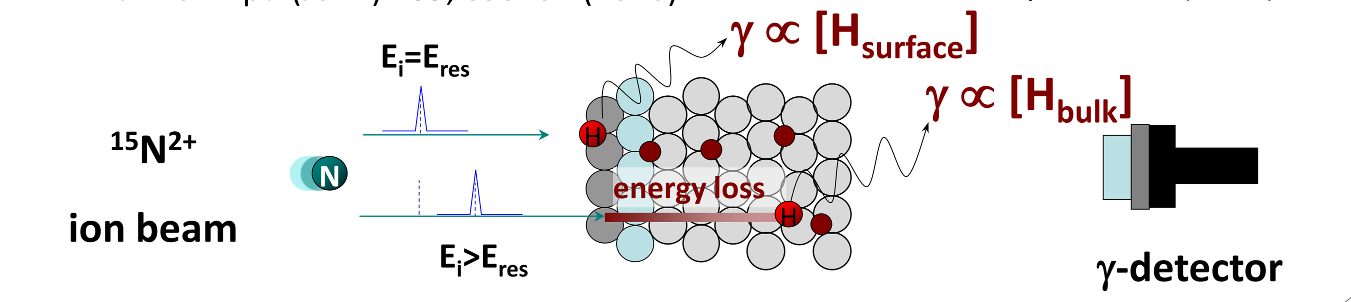
- ✓ *Nuclear Reaction Analysis (NRA)*: Quantitative ・ Non-destructive ・ High-resolution - Hydrogen Depth Profiling ・ Visualization of H-breathing by nanostructures ・ Depth-resolved H stability analysis (diffusion, desorption, reaction)
- ✓ *Thermal Desorption Spectroscopy (TDS)*: Bonding stability of H species ・ Hydrogen absorption kinetics ・ Isotope (D) labeling ・ Gas/surface/subsurface-H/D exchange mechanisms ・ Kinetic isotope effects

◆ Latest Research Topics

- ✓ *Hydrogen Storage & Retention* → H-Absorption/Release Mechanism
- ✓ *Hydrogenation Catalysis* → Reactivity of 'Subsurface-H'
- ✓ *(Photo)Catalysis* → H-Interactions & Diffusion in CeO_2 and TiO_2

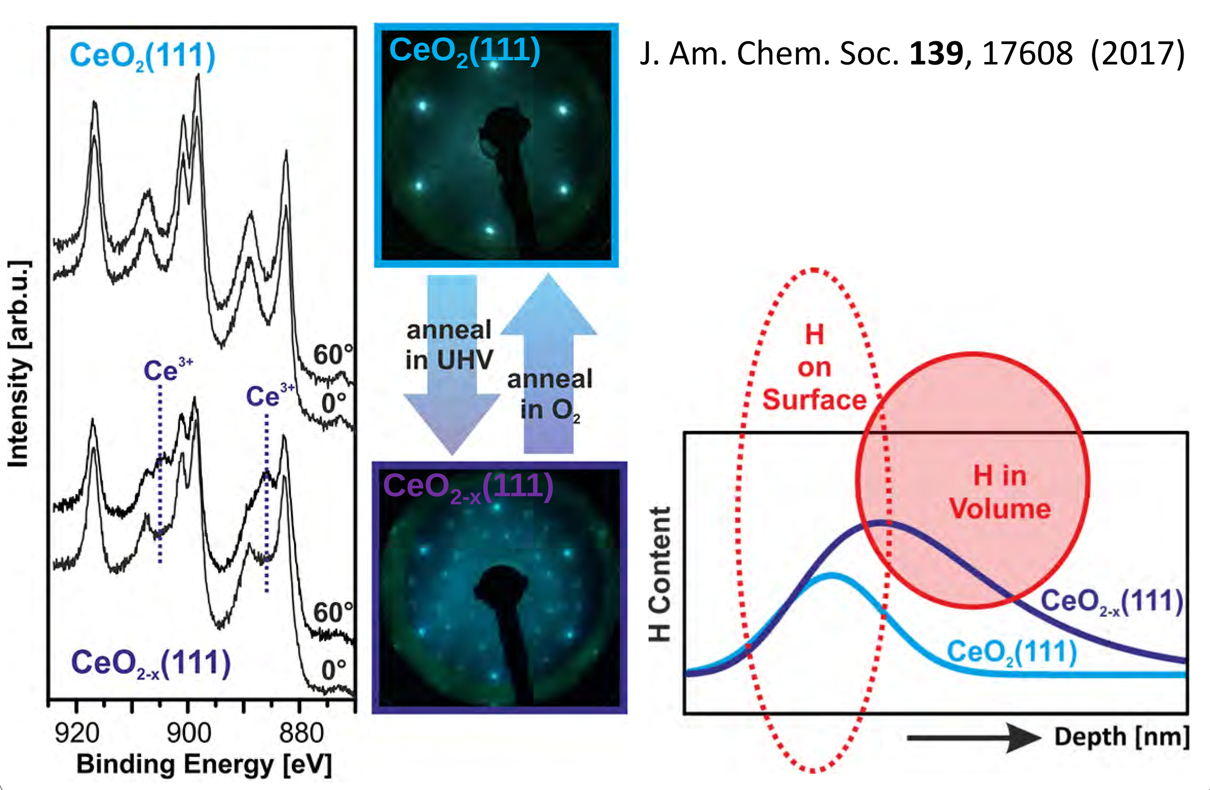
Principle of NRA $^{15}N(6.385\ MeV) + {}^{1}H \rightarrow {}^{12}C + a + \gamma(4.43\ MeV)$

J. Vis. Expt. (JoVE) 109, e53452 (2016)   Surf. Sci. Rep. 69, 196 (2014)



O-vacancies stabilize subsurface H in reduced CeO_{2-x}

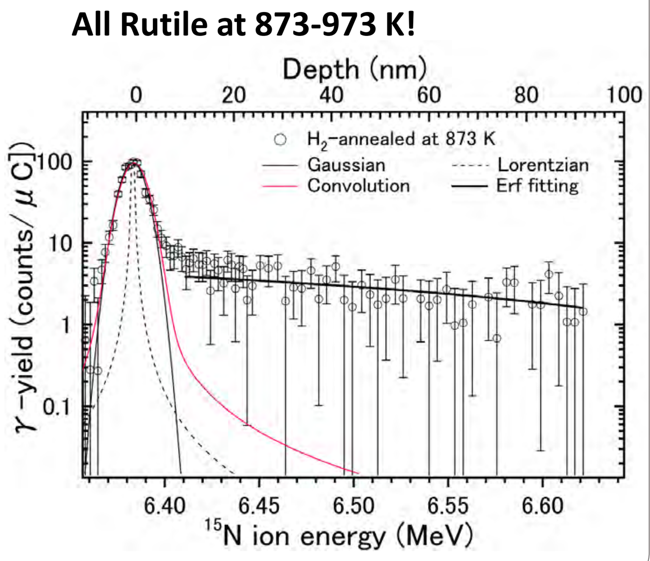
J. Am. Chem. Soc. 139, 17608 (2017)



Accelerated phase transition and H-diffusion in TiO_2 films

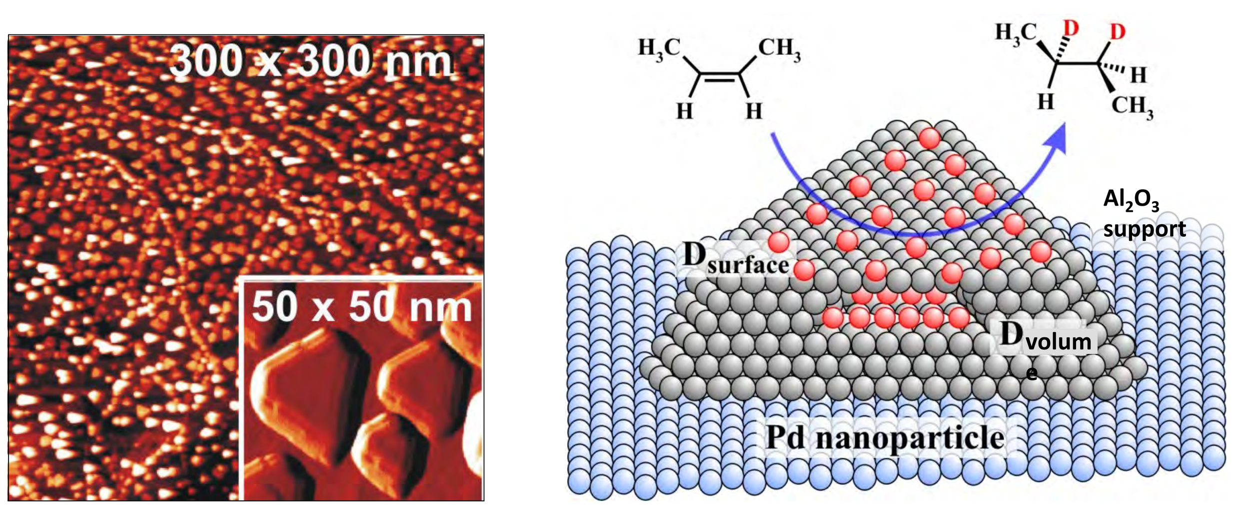
J. Phys. Chem. C 122, 23026 (2018)

H-diffusion profile in TiO_2 thin film: All Rutile at 873-973 K!



Hydrogen in Pd nanocrystals: Essential in hydrogenation catalysis

Angew. Chem. Int. Ed. 47, 9289 (2008)   Phys. Rev. B 77, 113412 (2008)

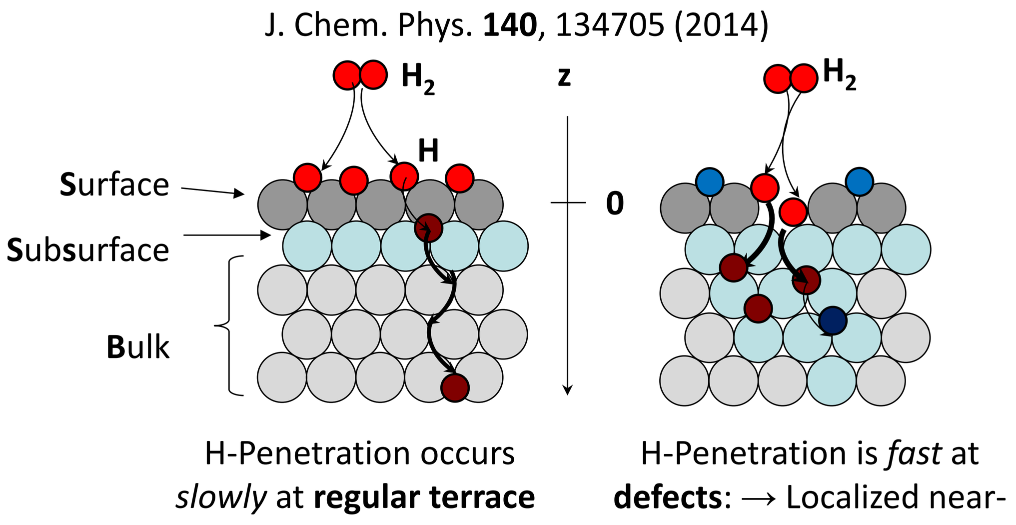


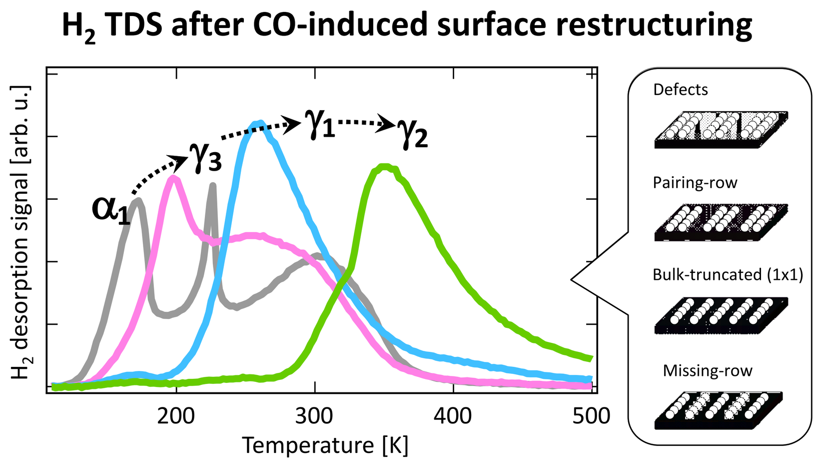
Model Hydrogenation Catalyst

=> Olefin (C=C) hydrogenation catalysis *requires* Pd-absorbed H !

Structure-sensitive H_2 absorption and H_2 desorption temperature control at Pd(110)

J. Chem. Phys. 140, 134705 (2014)   J. Phys. Chem. C 119, 11732 (2015)

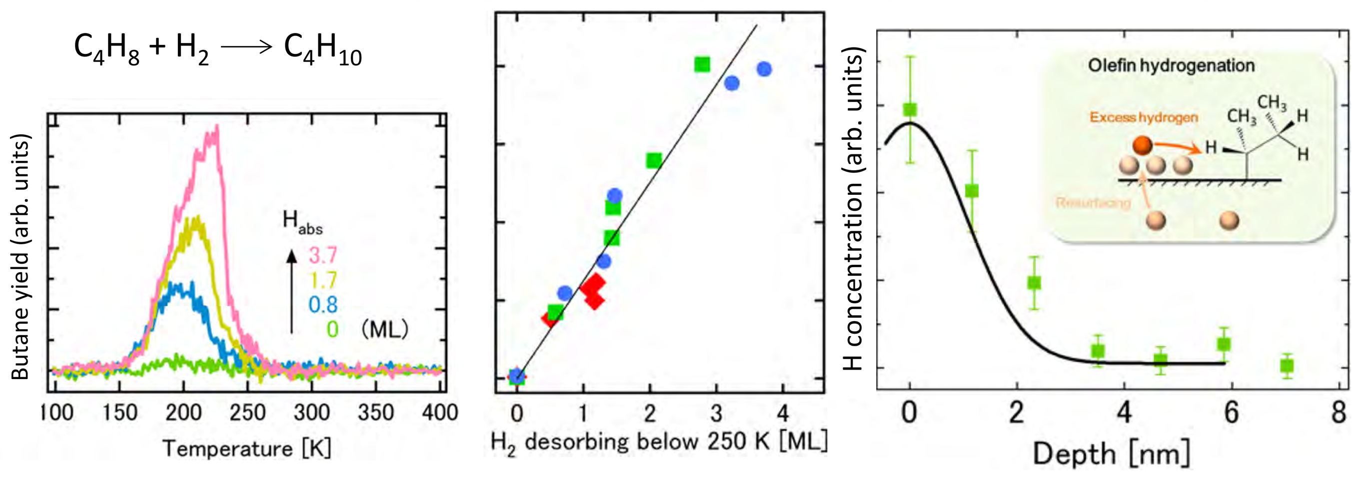




Resurfacing of Pd-absorbed H triggers hydrogenation catalysis

J. Phys. Chem. C 120, 11481 (2016)

Mechanism of olefin hydrogenation catalysis driven by Pd-dissolved hydrogen



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