<u>Researchgroup:</u>
Experimental X-ray studies of Liquids and Surfaces (XSoLaS)

The group focusses on x-ray scattering, x-ray spectroscopy and photoelectron spectroscopy techniques using synchrotron radiation sources and x-ray free electron lasers at facilities all around the world. The research is directed towards fundamental studies but related to challenges of societal importance. The research interests include chemical bonding and reactions on surfaces, ultrafast science, heterogeneous catalysis, electrocatalysis in fuel cells, photocatalysis for converting sunlight to fuels, structure, dynamics and physical properties of water and aqueous solutions.

The first X-ray Laser in the world: the Linac Coherent Lights Source at Stanford University

Contact:
Anders Nilsson
Professor
08-553787637

Research Projects:

Water

It is known since several decades that many properties of water such density, compressibility, heat capacity etc. become strongly anomalous as the temperature decreases. In particular, this deviation from a simple liquid behaviour becomes strongly enhanced as water is cooled below the freezing point into the metastable supercooled regime. Some of these properties seem to diverge around a mysterious temperature of 228 K and different theories have been proposed to explain this unusual behaviour. One of the most popular and that we are currently investigating with experimental techniques is the liquid-liquid transition (LLT) and liquid-liquid critical point (LLCP) model. Here we base our studies on using x-ray lasers to investigate deep supercooled water on timescales that is shorter than time for ice nucleation. It is also important to realize that the anomalous regime already occurs at much warmer temperatures even above room temperature. We are, therefore, also investigating water in the stable regime with an overall aim to establish a unified understanding of water from hot to deep supercooled conditions using synchrotron radiation. Here the hypothesis is that water at ambient temperature encompasses fluctuations around two local structures and that the dominating structure is a strongly distorted H-bonded environment but the tetrahedral local structure that are organised in small patches grows in population and size with decreasing temperature.

Same Contact with Anders Nilsson as above

Members:

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Postdoc

Fysikum

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Publications:

Origin of the Anomalous Properties in Supercooled Water Based on Experimental Probing Inside "No-Man's Land"
A. Nilsson
J. Non Cryst. Sol. X **14**, 100095 (2022).

Direct Observation of Ultrafast Hydrogen Bond Strengthening in Liquid Water

J. Yang, R. Dettori, J. P. F. Nunes, N. H. List, E. Biasin, M. Centurion, Z. Chen, A. A. Cordones, D. P. Deponte[1], T. F. Heinz, M. E. Kozina, K. Ledbetter, M-F Lin, A. M. Lindenberg, M. Mo, A. Nilsson, X. Shen, T. J. A. Wolf, D. Donadio, K. J. Gaffney, T. J. Martinez, X. Wang
Nature **596**, 531 (2021)

Enhancement and Maximum in the Isobaric Specific Heat Capacity Measurements of Deeply Supercooled Water using Ultrafast Calorimetry
H. Pathak, A. Späh, N. Esmaeildoost, J. A. Sellberg, F. Perakis, K. Amann-Winkel, M. Ladd Parada, J. Koliyadu, T. J. Lane, K. H. Kim, C. Yang, H. T. Lemke, R. A. Oggenfuss, P. Johnson, S. Zerdane, R. Mankowsky, P. Beaud and A. Nilsson.
Proceedings of the National Academy (USA) **118**, e2018379118 (2021).

Experimental Observation of the Liquid-Liquid Transition in Bulk Supercooled Water under Pressure
K. H. Kim, K. Amann-Winkel, N. Giovambattista, A. Späh, F. Perakis, H. Pathak, M. L. Parada, C Yang, D. Mariedahl, T. Eklund, T. J. Lane, S. You, S. Jeong, M. Weston, J. H. Lee, I. Eom, M. Kim, J. Park, S. H. Chun, P. H. Poole and A. Nilsson
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F. Perakis, G. Camisasca, T. J. Lane, A. Späh, K. T. Wikfeldt, J. A. Sellberg, F. Lehmkühler, H. Pathak, K. H. Kim, K. Amann-Winkel, S. Schreck, S. Song, T. Sato, M. Sikorski, A. Eilert, T. McQueen, H. Ogasawara, D. Nordlund, W. Roseker, J. Koralek, S. Nelson, P. Hart, R. Alonso-Mori, Y. P. Feng, D. L. Zhu, A. Robert, G. Grubel, L. G. M. PetterssonD. Pettersson, J. A. Sellberg, H. Pathak, F. Cavalca, D. Schlesinger, A. Ricci, A. Jain, B. Massani, F. Aubree, C. J. Benmore, T. Loerting, G. Grübel, L. G. M. Pettersson and A. Nilsson
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Maxima in the Thermodynamic Response and Correlation Functions of Deeply Supercooled Water
K. H. Kim, A. Späh, H. Pathak, F. Perakis, D. Mariedahl, K. Amann-Winkel, J. A. Sellberg, J. H. Lee, S. Kim, J. Park, K. H. Nam, T. Katayama and A. Nilsson
Science **358**, 1589 (2017).

Temperature-Independent Nuclear Quantum Effects on the Structure of Water
K. H. Kim, H. Pathak, A. Späh, F. Perakis, D. Mariedahl, J. A. Sellberg, T. Katayama, Y. Harada, H. Ogasawara, L. G.M. Pettersson, A. Nilsson
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F. Perakis, K. Amann-Winkel, F. Lehmkühler, M. Sprung, D. Pettersson, J. A. Sellberg, H. Pathak, A. Späh, F. Cavalca, D. Schlesinger, A. Ricci, A. Jain, B. Massani, F. Aubree, C. J. Benmore, T. Loerting, G. Grübel, L. G. M. Pettersson and A. Nilsson
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The Structural Origin of Anomalous Properties of Liquid Water
A. Nilsson and L. G. M. Pettersson
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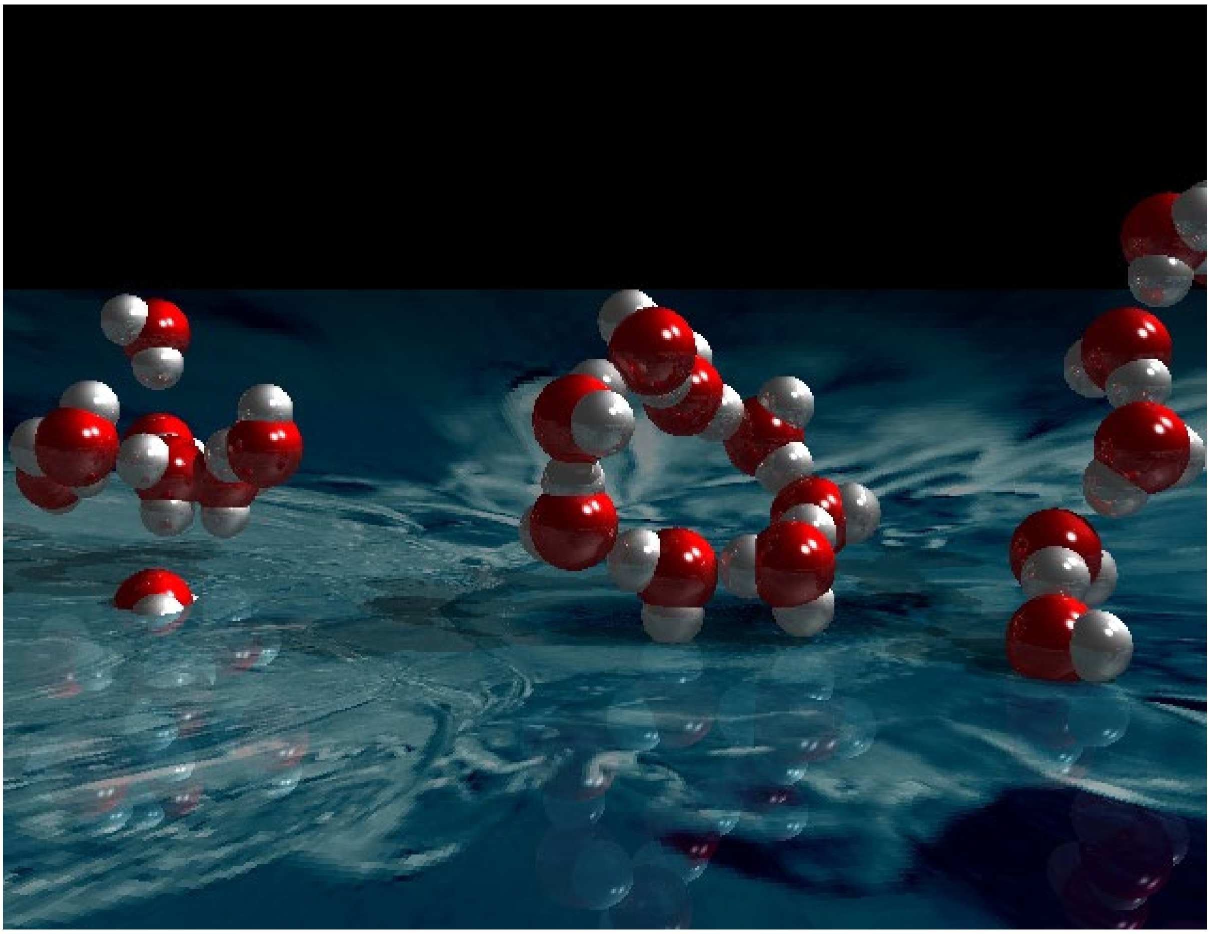
Ultrafast X-ray probing of water structure below the homogeneous ice nucleation temperature
J. A. Sellberg, C. Huang, T. A. McQueen, N. D. Loh, H. Laksmono, D. Schlesinger, R. G. Sierra, D. Nordlund, C. Y. Hampton, D. Starodub, D. P. DePonte, M. Beye, C. Chen, A. V. Martin, A. Barty, K. T. Wikfeldt, T. M. Weiss, C. Caronna, J. Feldkamp, L. B. Skinner, M. M. Seibert, M. Messerschmidt, G. J.Williams, S. Boutet, L. G. M. Pettersson, M. J. Bogan and A. Nilsson
Nature **510**, 381 (2014)



Different potential configurations of the hydrogen bonding network in water

Chemical Energy Transformations and Catalysis

There are many base chemicals of importance for the catalytic production of fertilizers, plastics, detergents, pharmaceuticals and fuels. Currently, the feedstock for the chemical industry is entirely based on fossil sources with an emittance of 8% of the worlds footprint of greenhouse gases (not including when fuels is being burned). There is an urgency to transform the chemical industry away from fossil sources and base it on captured CO_2 and H_2 from electrolysis of water. A future green transformation of the chemical industry, similar to the one with green steel, where wind or solar energy drives electrolytic cells is therefore a possibility. Catalysts are at the heart of these transformation currently based on fossil resources into products for use in everyday life. Naturally the catalysts in use today have been developed over a century with fossil resources in mind. In this research activity we suggest to further unravel the reaction mechanisms that enable novel catalysts to drive the reaction at radically lower pressures and temperatures that is better suited for a sustainable society. We are using a novel Stockholm built photoelectron spectroscopy instrument at the PETRA III synchrotron that operates at high pressures to measure the state of surface of various catalyst and molecular species on the surface at real reaction conditions. The goal is to understand the reaction induced dynamical changes of the catalyst and the mechanism. Typical reactions is Haber Bosch to produce ammonia from nitrogen and reduction of carbon dioxide to useful products mitigate climate change.

Contact Anders Nilsson (see above)

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Publications:

The state of zinc in methanol synthesis over a Zn/ZnO/Cu(211) model catalyst
P. Amann, B. Klötzer, D. Degerman, N. Köpfle, T. Götsch, P. Lömker, C. Rameshan, K. Ploner, D. Bikaljevic, H-Y. Wang, M. Soldemo, M. Shipilin, C. M. Goodwin, J. Gladh, J. H. Stenlid, M. Börner, C. Schlueter and A. Nilsson
Science **376**, 603 (2022).

In situ surface-sensitive investigation of multiple carbon phases on Fe(110) in Fischer-Tropsch synthesis

M. Shipilin, D. Degerman, P. Lömker, C. M. Goodwin, G. L. S. Rodrigues, M. Wagstaffe, J. Gladh, H. Y. Wang, A. Stierle, C. Schlueter, L. G.M. Pettersson, A. Nilsson, P. Amann
ACS Catal. **12**, 7609 (2022).

Operando Observation of Oxygenated Intermediates during CO Hydrogenation on Rh Single Crystals
D. Degerman, M. Shipilin, P. Lömker, C. M. Goodwin, S. M. Gericke, U. Hejral, J. Gladh, H-Y. Wang, C. Schlueter, A. Nilsson, and P. Amann
J. Am. Chem. Soc. **144,** 7038 (2022).

Direct Evidence of Subsurface Oxygen in Oxide-Derived Cu Catalyst for CO_2 Reduction by In-situ X-ray Photoelectron Spectroscopy
H-Y Wang, M. Soldemo, D. Degerman, P. Lömker, C. Schlueter, A. Nilsson, and P. Amann
Angew. Chem. In. Ed. **61,** e202111021 (2022).

The Structure of the Active Pd state During Catalytic CO oxidization
C. M. Goodwin, M. Shipilin, S. Albertin, U. Hejral, P. Lömker, H-Y. Wang, S. Blomberg, D. Degermann, C. Schlueter, A. Nilsson, E. Lundgren, P. Amann
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Key activity descriptors of nickel-iron oxygen evolution electrocatalysts in the presence of alkali metal cations
M. Görlin, J. H. Stenlid, S. Koroidov, H. Y. Wang, M. Börner, M. Shipilin, A. Kalinko, V. Murzin, O. V. Safonova, M. Nachtegaal, A. Uheida, J. Dutta, M. Bauer, A. Nilsson, O. Diaz-Morales
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P. Amann, D. Degerman, M. T. Lee, J. D. Alexander. M. Shipilin, H. Y. Wang, F. Cavalca, M. Weston, J. Gladh, M. Blom, M. Björkhage, P. Löfgren, C. Schlueter, P. Loemker, K. Ederer, W. Drube, H. Noei, J. Zehetner, H. Wentzel, J. Ålund and A. Nilsson
Rev. Sci. Instrum. **90**, 103102 (2019).

Operando XAS Study of the Surface Oxidation State on a Monolayer IrO_x on RuO_x and Ru Oxide Based Nanoparticles for Oxygen Evolution in Acidic Media
A. F. Pedersen, M. Escudero-Escribano, B. Sebok, A. Bodin, E. Paoli, R. Frydendal, D. Friebel, I. E. L. Stephens, J. Rossmeisl, I. Chorkendorff and A. Nilsson
J. Phys. Chem. B B **122**, 878 (2018)

Subsurface Oxygen in Oxide-Derived Copper Electrocatalysts for Carbon Dioxide Reduction
A. Eilert, F. Cavalca, F. S. Roberts, J. Osterwalder, C. Liu, M. Favaro, E. J. Crumlin, H. Ogasawara, D. Friebel, L. G. M. Pettersson, A Nilsson
J. Phys. Chem. Lett. **8**, 285 (2017)

Formation of Copper Catalysts for CO2 Reduction with High Ethylene/Methane Product Ratio Investigated with In-Situ X-ray Absorption Spectroscopy
A. Eilert, F. S. Roberts, D. Friebel and A. Nilsson
J. Phys. Chem. Lett. **7**, 1466 (2016)

Ambient-Pressure XPS Study of a Ni-Fe Electrocatalyst for the Oxygen Evolution Reaction
H. Ali-Löytty, M. W. Louie, M. R. Singh, L. Li, H. G. S. Casalongue, H. Ogasawara, E. J. Crumlin, Z. Liu, A. T. Bell, A. Nilsson, and D. Friebel
J. Phys. Chem. C **120,** 2247 (2016)

From the Sabatier Principle to a Predictive Theory of Transition-Metal Heterogeneous Catalysis

A. J. Medford, J. S. Hummelshøj, J. Voss, F. Abild-Pedersen, F. Studt, T. Bligaard, A. Nilsson, and J. K. Nørskov,
J. Catal. **328**, 26 (2015)

High Selectivity for Ethylene from Carbon Dioxide Reduction over Copper Nanocube Electrocatalysts

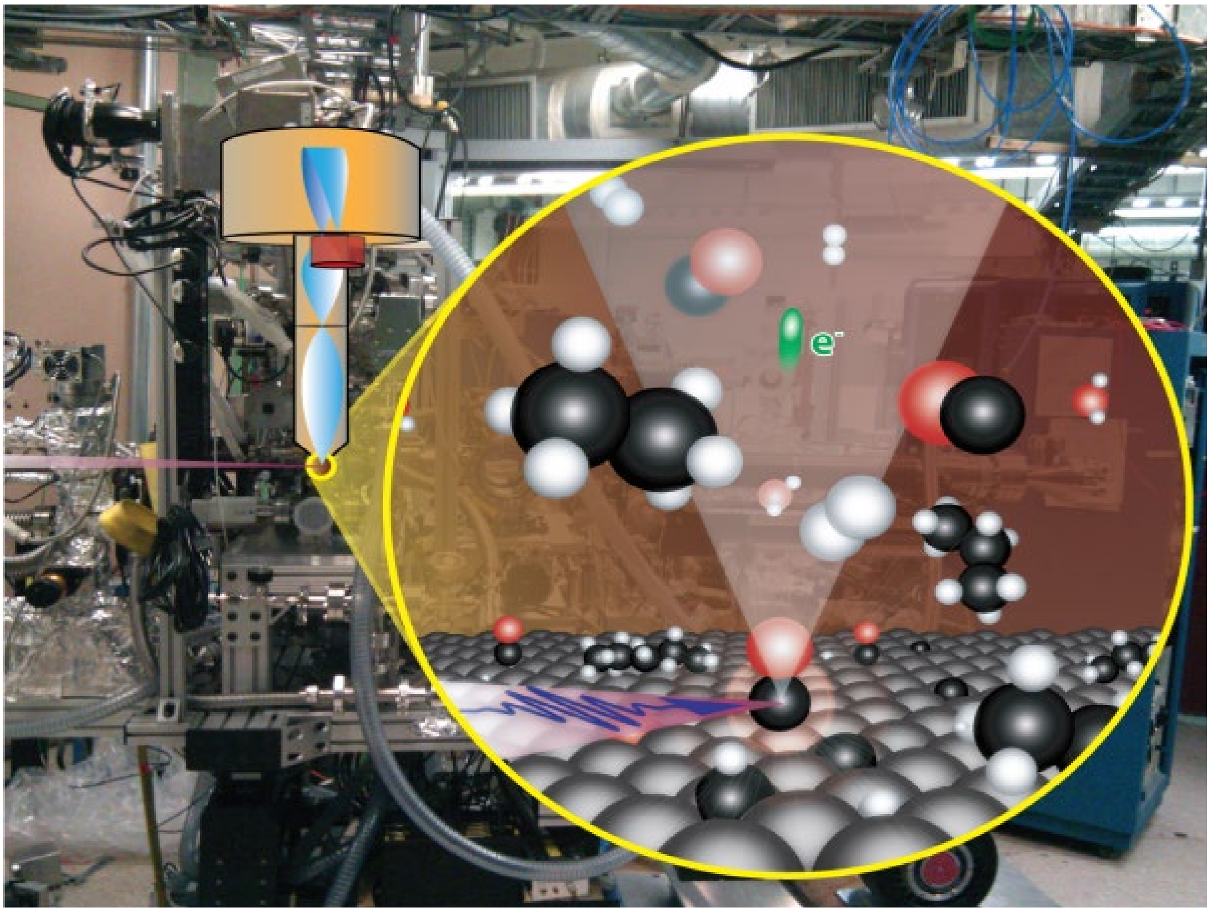
F**.** S. Roberts, K. P. Kuhl and A. Nilsson
Angew. Chem. Int. Ed **54**, 5179 (2015)

Identification of Highly Active Fe Sites in (Ni,Fe)OOH for Electrocatalytic Water Splitting
D. Friebel, M. W. Louie, M. Bajdich, K. E. Sanwald, Y. Cai, A. M. Wise, M. J. Cheng, D. Sokaras, T. C. Weng, R. Alonso-Mori, R. C. Davis, J. R. Bargar, J. K. Nørskov,
A. Nilsson, A. T. Bell
J. Am. Soc. **137**, 1305 (2015)

Mass-selected nanoparticles of PtxY as model catalysts for oxygen electroreduction
P. Hernandez-Fernandez, F. Masini, D. N.McCarthy, C. E. Strebel, D. Friebel, D. Deiana, P. Malacrida, A. Nierhoff, A. Bodin, A. M. Wise, J. H. Nielsen, T. W. Hansen, A. Nilsson, I. E. L. Stephens and I. Chorkendorff
Nat. Chem. **6,** 732 (2014).

In Situ Observation of Surface Species on Iridium Oxide Nanoparticles during the Oxygen Evolution Reaction
H. G. Sanchez Casalongue, M. L. Ng, S. Kaya, D. Friebel, H. Ogasawara and A. Nilsson
Angew. Chem. Int. Ed **53**, 1 (2014)

Direct observation of the oxygenated species during oxygen reduction reaction on a Pt fuel cell cathode
H. S. Casalongue, S. Kaya, V. Viswanathan, D. J. Miller, D. Friebel, H. A. Hansen, J. K. Nørskov, A. Nilsson and H. Ogasawara
Nat. Commun*.* **4,** 2817 (2013).



The differential pumped photoelectron spectroscopy system to probe the surface of model catalysts.

Ultrafast Surface Chemistry Probed with X-ray Lasers

Solid catalysts are frequently used to enhance the rate or selectivity of desirable chemical reactions. We aim to study the underlying mechanisms of these reactions by examining model catalytic systems. Often essential ingredients is to probe transient intermediates and molecular states close to the transition state where lifetimes are extremely short. It is therefore necessary to perform experiments using ultrafast techniques. To perform our experiments we use a laser “pump” pulse to provide energy to the system in a controlled manner with well-defined temporal characteristics. The energy from the laser pulse creates energetic electrons and phonons in the surface of the catalyst. Energy transfer to the adsorbed reactants can place the reactants into transiently excited states which could allow a reaction or transition to occur. X-ray “probe” pulses from an x-ray free electron laser with a controlled delay from the initial laser pulse can then be used to produce detectable photons or electrons from the system. The energy distribution of the particles produced will be a function of the initial and final electronic states of the reactants. By analysing the energy distribution of the emitted particles as a function of the pump-probe delay time we can measure changes in the electronic states of the reactants on ultrashort timescales. This can help to reveal short-lived intermediate states that may be necessary for a reaction to proceed, this information could be used to uncover the mechanisms behind certain reactions and to potentially design more efficient catalysts.

Contact Anders Nilsson

Collaboration with SLAC National Accelerator Laboratory and Stanford University

Publications:

Ultrafast adsorbate excitation probed with sub-ps resolution XAS
E. Diesen, H-Y. Wang, S. Schreck, M. Weston, H. Ogasawara, J. LaRue, F. Perakis, M. Dell'Angela, F. Capotondi, L. Giannessi, E. Pedersoli, D. Naumenko, I. Nikolov, L. Raimondi, C. Spezzani, M. Beye, F. Cavalca, B. Liu, J. Gladh, S. Koroidov, P. S. Miedema, R. Costantini, T. F. Heinz, F. Abild-Pedersen, J. Voss, A. C. Luntz, A. Nilsson
Phys. Rev. Lett. **127**, 016802 (2021).

Time-Resolved Observation of Transient Precursor State of CO on Ru(0001) using Carbon K-edge Spectroscopy
H. Y. Wang, S. Schreck, M. Weston, C. Liu, H. Ogasawara, J. LaRue, F. Perakis, M. Dell'Angelo, F. Capotondi, L. Giannessi, E. Pedersoli, D. Naumenko, I. Nikolov, L. Raimondi, C. Spezzani, M. Beye, F. Cavalca, B. Liu, J. Gladh, S. Koroidov, P. S. Miedema, R. Costantini, L. G. M. Pettersson and A. Nilsson
Phys. Chem. Chem. Phys. **22**, 2677 (2020).

Atom-Specific Activation in CO oxidation
S. Schreck, E. Diesen, J. LaRue, H. Ogasawara, K. Marks, D. Nordlund, M. Weston, M.Beye, F. Cavalca, F. Perakis, J. Sellberg, A. Eilert, K. H. Kim, G. Coslovich, R. Coffee, J. Krzywinski, A. Reid, S. Moeller, A. Lutman, H. Öström, L. G. M. Pettersson and A. Nilsson
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Real-Time Elucidation of Catalytic Pathways in CO Hydrogenation on Ru
J. LaRue, O. Krejč, L. Yu, M. Beye, M. L. Ng, H. Öberg, H. L. Xin, G. Mercurio, S. P. Moeller, J. Turner, D. Nordlund, R. Coffee, M. P. Minitti, W. Wurth, L. G. M Pettersson, H. Öström, A. Nilsson, F. Abild-Pedersen and H. Ogasawara
J. Phys. Chem. Lett. **8**, 3820 (2017)

Catalysis in Real Time using X-ray Lasers
A. Nilsson, J. LaRue, H. Öberg, H. Ogasawara, M. Dell'Angela, M. Beye, H. Öström, J. Gladh, J.K. Nørskov, W. Wurth, F. Abild-Pedersen and L.G.M. Pettersson
Chem. Phys. Lett. **675**, 145 (2017)

Chemical Bond Activation Observed with an X-ray Laser
M. Beye, H. Öberg, H. L. Xin, G. L. Dakovski, M. Dell'Angela, A. Föhlisch, J. Gladh, M. Hantschmann, F. Hieke, S. Kaya, D. Kühn, J. LaRue, G. Mercurio, M. P. Minitti, A. Mitra, S. P. Moeller, M. L. Ng, A. Nilsson, D. Nordlund, J. K. Nørskov, H. Öström, H. Ogasawara, M. Persson, W. F. Schlotter, J. A. Sellberg, M. Wolf, F. Abild-Pedersen, L. G. M Pettersson, W. Wurth
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J. LaRue, T. Katayama, A. Lindenberg, A. S. Fischer, H. Öström, A. Nilsson and H. Ogasawara Phys. Rev. Lett. **115**, 036103 (2015)

Probing the Transition State Region in Catalytic CO Oxidation on Ru
H. Öström, H. Öberg, H. Xin, J. LaRue, M. Beye, M. Dell'Angela, J. Gladh, M. L. Ng, J. A. Sellberg, S. Kaya, G. Mercurio, D. Nordlund, M. Hantschmann, F. Hieke, D. Kühn, W. F. Schlotter, G. L. Dakovski, J. J. Turner, M. P. Minitti, A. Mitra, S. P. Moeller, A. Föhlisch, M. Wolf, W. Wurth, M. Persson, J. K. Nørskov, F. Abild-Pedersen, H. Ogasawara, L. G. M Pettersson, A. Nilsson
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Real-Time Observation of Surface Bond Breaking with an X-ray Laser
M. Dell'Angela, T. Anniyev, M. Beye, R. Coffee, A. Föhlisch, J. Gladh, T. Katayama, S. Kaya, O. Krupin, A. Møgelhøj, D. Nordlund, J. K. Nørskov, H. Öberg, H. Ogasawara, H. Öström, L. G. M. Pettersson, W. F. Schlotter, J. A. Sellberg, F. Sorgenfrei, J. Turner, M. Wolf, W. Wurth, A. Nilsson
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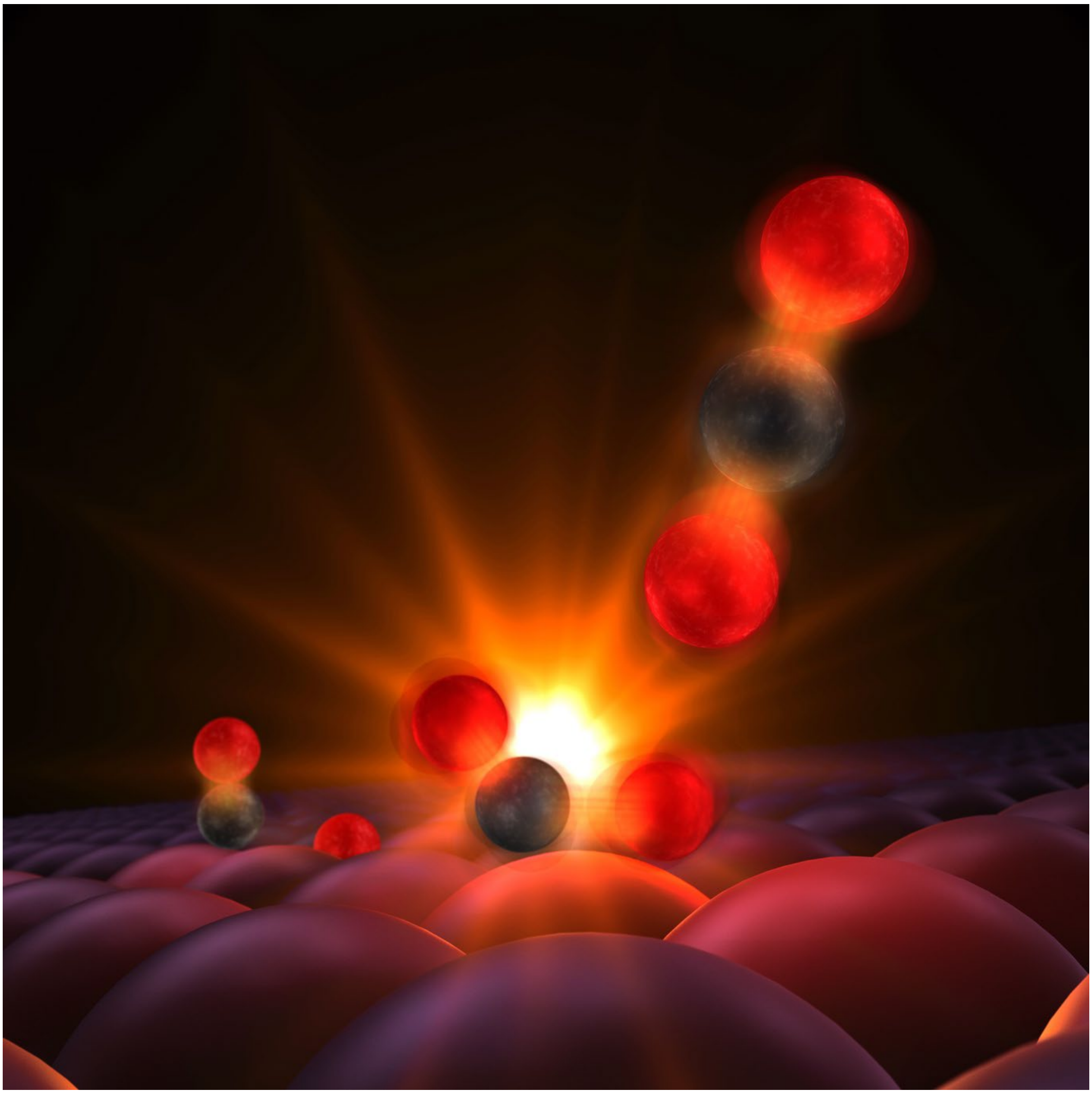

The transition state during CO oxidation on a surface