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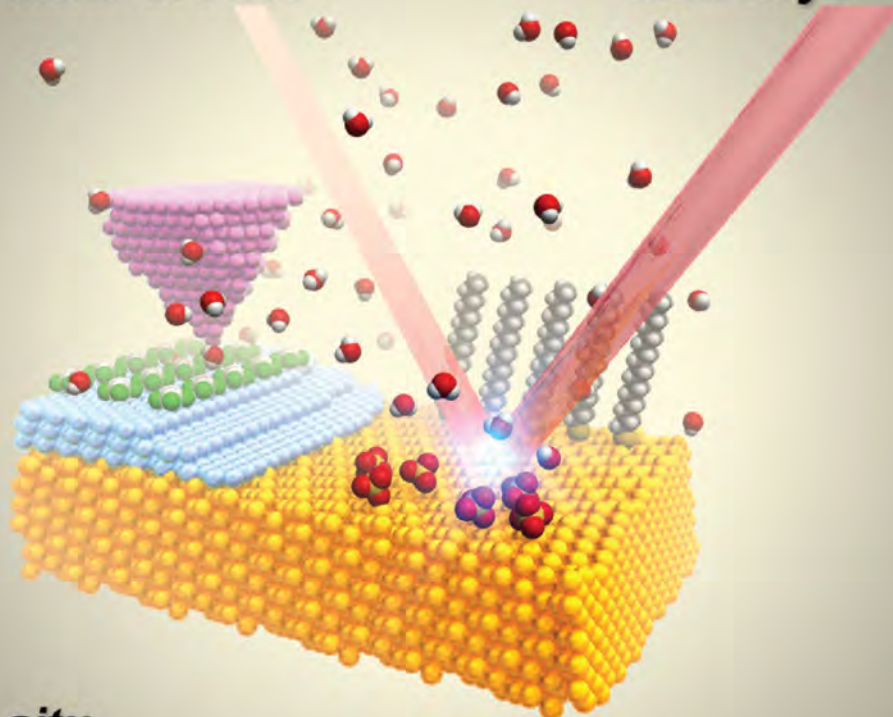
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# THE JOURNAL OF PHYSICAL CHEMISTRY

C

*Electrochemical  
Epitaxial Growth*

*Self-Assembled  
Monolayer*



*In situ  
Characterization  
Methods*

*Photoinduced  
Electron Transfer*

Electrochemistry of  
Ordered Interfaces -  
Design, Construction,  
and Interrogation  
of Functional  
Electrochemical  
Interphases with  
Atomic/Molecular  
Resolution

KOHEI UOSAKI Festschrift



## Preface to the Kohei Uosaki Festschrift: Electrochemistry of Ordered Interfaces—Design, Construction, and Interrogation of Functional Electrochemical Interphases with Atomic/Molecular Resolution



This issue of the *Journal of Physical Chemistry C* is dedicated to Kohei Uosaki, Fellow of National Institute for Materials Science (NIMS) and Adjunct Professor of Graduate School of Chemical Sciences and Engineering and Professor Emeritus of Hokkaido University. This collection of articles from many of his colleagues, collaborators, friends, and students is contributed in recognition of Uosaki's many outstanding scientific accomplishments and especially in honor of his contributions toward Surface Physical Chemistry in general and Electrochemical Surface Science in particular.

Kohei Uosaki was born and raised in Kobe, Japan. He received his B. Eng. and M. Eng. degrees in Applied Chemistry from Osaka University in 1969 and 1971, respectively, with thesis work on "Ion-solvent Interactions and its Effect on Nucleophilic Reactions" in the Organic Chemistry Laboratory led by Professor Niichiro Tokura in collaboration with Dr. Toshihiko Kondo, Dr. Takeki Matsui, Mr. Masaru Onishi, and Mr. Sho-ichiro Takezawa. Immediately after the completion of his M. Eng., Uosaki joined Mitsubishi Petrochemical Co. Ltd. as a Research Chemist where he worked on "Ion-exchange Membrane and its Application to Desalination Process by Electrodialysis" with Mr. Katsuo Akaiwa, Mr. Taiko Uno, and Mr. Naotoshi Watanabe. Uosaki was responsible for the design of a pilot desalination plant to be used in the Kashima Petrochemical Complex of Mitsubishi Petrochemical Co. Ltd. in 1973/1974.

He was given a leave of absence by the company to carry out his Ph.D. studies at the School of Physical Sciences of Flinders University in South Australia in 1974 under the supervision Prof. John O'M Bockris. This step marked the start of his career in electrochemistry. Uosaki investigated photoelectrochemical hydrogen evolution at single-crystal p-type semiconductors and correlated their photoelectrochemical characteristics with physical properties of the semiconductors. His work emphasized the importance of the potential drop in the double layer formed at the semiconductor-electrolyte interface and the charge transfer step at this interface, features which were

initially neglected but later accepted. After a very tough two and half years, involving numerous and extensive discussions with Prof. Bockris, Uosaki completed his Ph.D., the topic of which addressed theoretical and experimental studies on photoelectrochemical production of hydrogen, and returned to Japan in 1976. He worked at Mitsubishi Petrochemical for two more years examining the electrochemistry of viologens for electrochromic displays and household applications of solar thermal energy with Dr. Masaru Fuwa and Mr. Michihiro Saito. Uosaki started his academic career in 1978 as a Research Officer, a postdoctoral position, in the Inorganic Chemistry Laboratory at Oxford University where he worked on the electrochemistry of cytochrome-c in collaboration with Prof. H. Allen O. Hill, FRS.

After one and a half years in Oxford, Uosaki was appointed Assistant Professor in the Department of Chemistry, Faculty of Science, Hokkaido University in 1980, and was promoted to Associate Professor in the same department in 1981. Here, he initiated new work examining electrochemical semiconductor interfaces for the production of solar fuels. He investigated the effects of surface modification by metals and metal ions and showed they may act as surface recombination centers and/or catalysts. This work was summarized in an important review entitled "Theoretical Aspects of Semiconductor Electrochemistry" in Volume 18 of the Modern Aspects of Electrochemistry series, which was published in 1986. Uosaki also made breakthroughs in the electrochemical deposition of CdSe and CdSe<sub>x</sub>Te<sub>1-x</sub>. He quickly realized the importance of evaluating the electrode/electrolyte interface in situ in real time by using nonelectrochemical techniques. In 1982, Uosaki spent 3 months in Prof. Art Ellis's laboratory at the University of Wisconsin—Madison to learn electro- and photoluminescence methods to probe the semiconductor electrode/electrolyte interface. Uosaki utilized these luminescent techniques and later developed many other methods to probe the structure of the electrode surface in situ. He was one of the pioneers applying scanning probe microscopy (SPM), i.e., STM and AFM, to electrochemistry and was an invited speaker at the Symposium on "Tunneling Microscopy and Related Techniques" at the 174th Meeting of the Electrochemical Society (ECS) in 1988, the first symposium on SPM at an ECS meeting. While most SPM studies at that time were limited to examining under potential deposition (UPD) on single-crystal surfaces, he investigated more dynamic systems such as those involving metal deposition and dissolution.

In 1990, Uosaki was appointed Professor of Physical Chemistry in the Department of Chemistry, Faculty of Science, Hokkaido University, the chair once held by Prof. Juro Horiuchi. In the Physical Chemistry Laboratory, Uosaki and

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his team members explored important electrochemical and photoelectrochemical phenomena by using ultrasensitive in situ spectroscopic techniques as well as through the use of synthesis to construct ordered, efficient electron transfer systems. Being aware of the limitations of SPM, he employed complementary in situ techniques, such as surface X-ray scattering (SXS) and X-ray absorption fine structure (XAFS). Using these techniques, Uosaki showed that Pd deposition on Au(111) and Au(100) proceeded in a layer-by-layer fashion with an adsorbed Pd complex on the surface, giving a pseudomorphic Pd structure with unique electrocatalytic activity. Combining AFM, XAFS, and light scattering, Uosaki clarified the mechanism of Cu deposition on GaAs from the very initial stage to multilayer deposits, a truly unique feat. In addition to his spectroelectrochemical emphasis, Uosaki's main contributions are in the area of self-assembled monolayers (SAMs) of thiol molecules on gold with various functionalities. He synthesized novel thiol molecules and formed SAMs with exhibiting functions such as redox, proton-coupled redox, electrochemically generated luminescence, second harmonic generation, and photoinduced electron transfer. In a particularly important finding, he demonstrated more than 10% quantum efficiency for photoinduced uphill electron transfer at a gold electrode modified with a ferrocene-porphyrin-thiol SAM, the highest efficiency obtained from a SAM modified metal electrode at that time.

During 1997–2000, Uosaki led a project entitled “Electrochemistry of Ordered Interfaces” a Priority Area Research of the Grants-in-Aid program. The project consisted of 80 leading Japanese electrochemists, supported by Ministry of Education, Culture, Science, Sports, and Technology, Japan. The ultimate goal of this project was to understand and control electrode reactions at the atomic and molecular levels. Each member belonged to one of four research groups: “Interfacial Structures and Electron Transfer Dynamics”, “Fabrication of Novel Interfaces by Using Electrochemical Techniques and Development of High Performance Batteries”, “Ordered Functional Interfaces for Molecular Recognition and Sensing”, and “Photoelectrode Reactions and Photofunctional Interfaces”. Collections of the achievements of this project were published as three special issues on “Electrochemistry of Ordered Interfaces” in *The Journal of Electroanalytical Chemistry* (Sept. 1999), *Electrochemistry* (Dec. 1999), and *Physical Chemistry and Chemical Physics* (Aug. 2001). Many special symposia related to “Electrochemistry of Ordered Interfaces”, both domestic and international, were organized, including a series of symposia in the Spring/Fall Meetings of the Electrochemical Society of Japan and Chemical Society of Japan and the International Symposium on Electrochemistry of Ordered Interfaces in 1998 and 2000.

Uosaki and his group continued to explore “Electrochemistry of Ordered Interfaces - design, construction, interrogation, and applications of functional electrochemical interphases” after this project was completed. He and his colleagues improved the efficiency of photoinduced uphill electron transfer at a gold electrode modified with a ferrocene-porphyrin-thiol SAM by placing a gold nanocluster on top of the SAM thereby utilizing a plasmon effect. They also utilized this combination to obtain Raman spectra of monolayers on well-defined surfaces. Uosaki also extended his study of SAMs of thiol molecules on gold to mono- and multilayers of organic molecules directly bonded to hydrogen-terminated Si(111). He is also a forerunner in applying nonlinear optical spectroscopy, in particular second

harmonic generation (SHG) and sum frequency generation (SFG) spectroscopy, to electrochemical interfaces. Uosaki used SHG to follow Te and Pd electrodeposition and to probe surface-confined CO on the surface during the anodic oxidation of HCHO. He also demonstrated that surface chirality can be evaluated by SHG and used SFG to investigate the structure of water at electrode surfaces, one of the most important issues in electrochemistry, showing that interfacial water structure strongly depends on the nature of the electrode and surface charge.

After he retired from Hokkaido University in March 2010, Uosaki joined the National Institute for Materials Science (NIMS) as a Principal Investigator of the International Center for Materials Nanoarchitectonics (MANA) and was appointed a NIMS Fellow in 2013. There, he continued cutting edge electrochemical surface science research. His group demonstrated that a Si surface modified with mono/multilayers of viologen incorporating a Pt complex exhibits high photoelectrochemical hydrogen evolution efficiency and proposed that the Pt complex acted as a “confined molecular catalyst”. His group also developed two important techniques to determine electronic structure at the electrode/electrolyte interface in situ. One of these is an electrochemical XPS system in which hard X-rays and a very thin Si window are utilized to probe the electronic structure of the electrochemical interface in ordinary solvents. The other technique uses a potential-dependent double resonance (DR-) sum frequency generation (SFG) system. DR-SFG uses both tunable visible and infrared light, and the SFG signal reflects not only the resonance of the IR with an interfacial vibrational state but also that of visible light with an interfacial electronic state, the nature of which is also a function of potential. His group also proposed and experimentally proved that an insulating BN nanosheet on an electrocatalytically inert gold substrate acts as an electrocatalyst for oxygen reduction reaction (ORR), a key reaction in fuel cells. These examples demonstrate that Uosaki is still a significant participant in electrochemical surface science research.

Uosaki's scientific achievements have been recognized widely by many prizes including the Scientific Achievement Award of the Electrochemical Society of Japan in 1993, the Chemical Society of Japan Award in 2010, and the Surface Science Society of Japan Award in 2014. He is a Fellow of IUPAC, the Royal Society of Chemistry, the International Society of Electrochemistry, the Electrochemical Society, and the Chemical Society of Japan. He was recognized by his alma mater, Flinders University, with a Distinguished Alumni Award in 2006 and was admitted to the Degree of Doctor of Science *honoris causa* in 2015. Prof. Uosaki has contributed to the physical chemistry and electrochemistry communities not only in scientific research but also as a member of editorial boards of many scientific journals including *Chemistry Letters*, *Bulletin of the Chemical Society of Japan*, *Electrochemistry Communications*, *Journal of Electroanalytical Chemistry*, and *Physical Chemistry and Chemical Physics* and in organizing many international conferences and symposia.

During his academic career, he mentored 36 Ph. D. (including 2 current students), 50 Master, and 37 undergraduate students and 36 postdocs, most of whom are now actively working as professional scientists/engineers in universities, public institutions, and industry worldwide. He has also accepted many visiting scientists and students from both domestic and foreign institutions to his laboratory and

played important roles establishing international collaborative relations between his institutions, Hokkaido University and NIMS, with various universities in China, Korea, and Australia.

It is a great honor for us to be included among friends, colleagues, and students of Kohei Uosaki. It is with great pleasure that we dedicate this special issue of *The Journal of Physical Chemistry C* to him.

**Andrew A. Gewirth**

University of Illinois

**Juan M. Feliu**

Universidad de Alicante

**Susumu Kuwabata**

Osaka University

**Kei Murakoshi**

Hokkaido University

## Curriculum Vitae of Kohei Uosaki

### ■ BIRTH DATE AND PLACE

February 1, 1947 in Kobe, Japan

### ■ PRESENT POSITIONS

#### National Institute for Materials Science (NIMS).

Fellow

Executive Advisor to the President

Director, Center for Green Research on Energy and Environmental Materials

Director, Global Research Center for Environment and Energy based on Nanomaterials Science (GREEN)

Principal Investigator, International Center for Materials Nanoarchitectonics (MANA)

#### Hokkaido University.

Professor Emeritus

Adjunct Professor, Graduate School of Chemical Sciences and Engineering

#### Japan Science and Technology Agency (JST).

Special Fellow, Center for Research and Development Strategy (CRDS)

Program Officer, Specially Promoted Research on Innovative Next Generation Battery, Advanced Low Carbon Technology R & D Program (ALCA-SPRING)

### ■ EDUCATION

- 1965.4–1969.3: Department of Applied Chemistry, Faculty of Engineering, Osaka University: B. Eng. 1969
- 1969.4–1971.3: Division of Applied Chemistry, Graduate School of Engineering, Osaka University: M. Eng. 1971
- 1974.4–1976.10: School of Physical Sciences, The Flinders University of South Australia: Ph. D. 1977

### ■ PROFESSIONAL EXPERIENCES

- 1971.4–1978.10: Research Chemist, Mitsubishi Petrochemical Co. Ltd.
- 1978.10–1980.3: Research Officer (Post doctoral Fellow), Inorganic Chemistry Laboratory, Oxford University
- 1980.4–1981.10: Assistant Professor, Department of Chemistry, Faculty of Science, Hokkaido University
- 1981.10–1990.8: Associate Professor, Department of Chemistry, Faculty of Science, Hokkaido University
- 1990.9–2010.3: Professor of Physical Chemistry, Department of Chemistry, Faculty of Science, Hokkaido University
- 1993.4–1994.3: Chairman, Chemistry Department, Hokkaido University
- 1999.4–2000.3: Advisor to the President, Hokkaido University
- 2000.4–2002.3: Director, Catalysis Research Center, Hokkaido University
- 2000.4–2002.3: Senate, Hokkaido University

- 2005.4–2006.3: Chairman, Division of Chemistry, Hokkaido University
- 2007.9–2010.3: Director, Collaborative Research Center for Molecular and System Life Sciences, Hokkaido University
- 2008.10–2010.3: Director, Collaborative Research Center for Strategic Utilization of Elements, Hokkaido University
- 2010.4–present: Principal Investigator, MANA, NIMS
- 2013.2–present: Fellow, NIMS
- 2013.2–present: Director, GREEN, NIMS
- 2016.4–present: Director, Center for Green Research on Energy and Environmental Materials, NIMS

### ■ CONCURRENT APPOINTMENT

- 2006–present: Associate Member, Science Council of Japan
- 2008.10–2010.3: Satellite Principal Investigator and Coordinator of Nanogreen Field, MANA, NIMS
- 2009.4–present: Special Fellow, Nanotechnology and Materials, CRDS, JST
- 2010.4–present: Adjunct Professor, Graduate School of Chemical Sciences and Engineering, Hokkaido University
- 2012.7–2016.6: Technical Advisor, Intellectual Property High Court
- 2013.8–present: Program Officer of Specially Promoted Research for Innovative Next Generation Battery, Advanced Low Carbon Technology R & D Program (ALCA-SPRING), JST

### ■ VISITING PROFESSORSHIP, ETC.

- 1982.9–1982.12: Visiting Associate Professor, Department of Chemistry, University of Wisconsin—Madison
- 1989.7–1989.9: JSPS (Japan Society for Promotion of Science) Visiting Scientist, Department of Physical Chemistry, Gothenburg University (Sweden)
- 1994.12: Visiting Professor, Graduate School of Engineering, Osaka University
- 1995.9: JSPS Visiting Scientist, Department of Chemistry, Oxford University
- 1995.10: Visiting Scientist, National Institute of Biosciences and Human-Technology, Agency of Industrial Science and Technology
- 1997.4–2014.3: Research Advisor, Japan Atomic Energy Research Institute
- 2004.8–2004.9: JSPS Visiting Scientist, Department of Chemistry, Vienna Institute of Technology
- 1997.6: Visiting Professor, Graduate School of Engineering, Yamanashi University

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- 1998.6: Visiting Professor, Graduate School of Science, Nagoya University
- 1998.1: Visiting Professor, Graduate School of Engineering Science, Osaka University
- 1999.5: Visiting Professor, Graduate School of Engineering, Tokyo University
- 1999.11: Visiting Professor, Graduate School of Engineering, Osaka University
- 2000.6: Visiting Professor, Graduate School of Science, Tohoku University
- 2004.1: Visiting Professor, Institute of Multidisciplinary Research for Advanced Materials, Tohoku University
- 2005.10–2011.3: Guest Scientist, RIKEN
- 2007.3–present: Concurrent Professor, College of Chemistry and Chemical Engineering, Nanjing University (China)
- 2007.6: Visiting Professor, Graduate School of Science, Kyushu University
- 2010.11–2010.12: Visiting Professor, Graduate School of Humanities and Sciences, Ochanomizu University
- 1999–2009: Regional Editor for Asia–Pacific, Electrochemistry Communications
- 1999–2007: Editorial Board, Journal of Electroanalytical Chemistry
- 1999–2007: International Advisory Editorial Board, Physical Chemistry and Chemical Physics (The Royal Society of Chemistry)
- 2009–present: Editorial Board, Electrochemistry Communications

#### ■ COMMITTEE MEMBERS OF GOVERNMENT AND RELATED ORGANIZATIONS

- 1997–2005: Member, Research Liaison Committee for Chemistry, Science Council of Japan
- 2001–2011: Member, Committee on Nanotechnology and Materials, Ministry of Education, Culture, Sports, Science and Technology (MEXT)
- 2006–2011: Advisor, PRESTO “Search for Nonmanufacturing Technology and Its Development”, Japan Science and Technology Agency (JST)
- 2011–2015: Chairman, Technical Committee for Carbon Alloy Catalyst for Fuel Cell, New Energy and Industrial Technology Development Agency (NEDO)
- 2011–present: Member, Promotion Committee, Advanced Low Carbon Technology Research and Development Program (ALCA), JST
- 2011–present: Advisor, PRESTO “Energy Conversion Interface”, JST
- 2013–present: Member, Committee for Energy Strategy, Council for Science, Technology and Innovation, Cabinet Office
- 2013–present: Member, Selection and Evaluation Committee (Materials), Program for Leading Graduate Schools, JSPS
- 2014–present: Member, Joint Committee between MEXT and METI (Ministry of Economy, Trade and Industry) for Energy and Environment
- 2015–2016: Member, Committee for Intellectual Property of University, MEXT

#### ■ EDITORSHIP

- 1990–1993: Associate Editor, Chemistry Letters (The Chemical Society of Japan)
- 1993–1995: Associate Editor, Chemistry and Education (The Chemical Society of Japan)
- 1993–1996: Associate Editor, Catalysis (Catalysis Society, Japan)
- 1996–1999: Associate Editor, Bulletin of Chemical Society of Japan

## Awards and Honors to Kohei Uosaki

- (1) April, 1993; **Scientific Achievement Award**, The Electrochemical Society of Japan
- (2) April, 2000; **Best Paper Award**, The Electrochemical Society of Japan
- (3) April, 2002; **Fellow**, International Union of Pure and Applied Chemistry
- (4) May, 2005; **Fellow**, The Royal Society of Chemistry (UK)
- (5) March, 2006; **Distinguished Alumni Award**, Flinders University
- (6) November, 2008; **Fellow**, The International Society of Electrochemistry
- (7) October, 2009; **Fellow**, The Electrochemical Society
- (8) October, 2009; **BCSJ Award**, The Chemical Society of Japan
- (9) March, 2010; **Chemical Society of Japan Award**, The Chemical Society of Japan
- (10) April, 2010; **Professor Emeritus**, Hokkaido University
- (11) October, 2009; **Special Lectureship Award**, The Japanese Photochemistry Association
- (12) May, 2014; **Society Award**, The Surface Science Society of Japan
- (13) September, 2015; **Degree of Doctor of Science *honoris causa***, Flinders University
- (14) March, 2016; **Fellow**, The Chemical Society of Japan

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