

The public reporting burden for this collection of information is estimated to average 1 hour per response, including the time for reviewing instructions, searching existing data sources, gathering and maintaining the data needed, and completing and reviewing the collection of information. Send comments regarding this burden estimate or any other aspect of this collection of information, including suggestions for reducing this burden, to Washington Headquarters Services, Directorate for Information Operations and Reports, 1215 Jefferson Davis Highway, Suite 1204, Arlington VA, 22202-4302. Respondents should be aware that notwithstanding any other provision of law, no person shall be subject to any penalty for failing to comply with a collection of information if it does not display a currently valid OMB control number.
PLEASE DO NOT RETURN YOUR FORM TO THE ABOVE ADDRESS.

1. REPORT DATE (DD-MM-YYYY) 06-08-2021	2. REPORT TYPE Final Report	3. DATES COVERED (From - To) 1-Oct-2017 - 31-May-2021
---	--------------------------------	--

4. TITLE AND SUBTITLE Final Report: Super-resolution Imaging of Surface-Plasmon Enhanced Catalysis at Nanoscale Gaps	5a. CONTRACT NUMBER W911NF-17-1-0590
	5b. GRANT NUMBER
	5c. PROGRAM ELEMENT NUMBER 611102

6. AUTHORS	5d. PROJECT NUMBER
	5e. TASK NUMBER
	5f. WORK UNIT NUMBER

7. PERFORMING ORGANIZATION NAMES AND ADDRESSES Cornell University Office of Sponsored Programs 373 Pine Tree Road Ithaca, NY 14850 -2820	8. PERFORMING ORGANIZATION REPORT NUMBER
--	--

9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS (ES) U.S. Army Research Office P.O. Box 12211 Research Triangle Park, NC 27709-2211	10. SPONSOR/MONITOR'S ACRONYM(S) ARO
	11. SPONSOR/MONITOR'S REPORT NUMBER(S) 70743-CH.11

12. DISTRIBUTION AVAILABILITY STATEMENT Approved for public release; distribution is unlimited.
--

13. SUPPLEMENTARY NOTES The views, opinions and/or findings contained in this report are those of the author(s) and should not be construed as an official Department of the Army position, policy or decision, unless so designated by other documentation.

14. ABSTRACT

15. SUBJECT TERMS

16. SECURITY CLASSIFICATION OF:	17. LIMITATION OF ABSTRACT	15. NUMBER OF PAGES	19a. NAME OF RESPONSIBLE PERSON Peng Chen
a. REPORT UU	b. ABSTRACT UU	c. THIS PAGE UU	19b. TELEPHONE NUMBER 607-254-8533

RPPR Final Report
as of 15-Oct-2021

Agency Code: 21XD

Proposal Number: 70743CH

Agreement Number: W911NF-17-1-0590

INVESTIGATOR(S):

Name: Peng Chen
Email: pc252@cornell.edu
Phone Number: 6072548533
Principal: Y

Organization: **Cornell University**

Address: Office of Sponsored Programs, Ithaca, NY 148502820

Country: USA

DUNS Number: 872612445

EIN: 150532082

Report Date: 31-Aug-2021

Date Received: 06-Aug-2021

Final Report for Period Beginning 01-Oct-2017 and Ending 31-May-2021

Title: Super-resolution Imaging of Surface-Plasmon Enhanced Catalysis at Nanoscale Gaps

Begin Performance Period: 01-Oct-2017

End Performance Period: 31-May-2021

Report Term: 0-Other

Submitted By: Peng Chen

Email: pc252@cornell.edu

Phone: (607) 254-8533

Distribution Statement: 1-Approved for public release; distribution is unlimited.

STEM Degrees: 3

STEM Participants: 4

Major Goals: Please see the uploaded pdf file.

Accomplishments: Please see the uploaded pdf file.

Training Opportunities: The research over the entire funding period has provided training for a number of postdocs: Dr. Ming Zhao, Dr. Wenjie Li, Dr. Xiaowen Mao, Dr. Youngchan Park, Dr. Kushal Sengupta, as well as Dr. Rong (Rocky) Ye.

It provided training for a graduate student, Udit Chakraborty, and a MS student: Heting Pu, for her Master thesis research.

It also provided training for two summer REU students: Zhaohong Wang from Shanghai JiaoTong University, China, and Diego A. Garzon from Universidad de los Andes, Colombia.

Results Dissemination: Please see the uploaded pdf file.

Honors and Awards: Please see uploaded pdf file.

Protocol Activity Status:

Technology Transfer: 1. Peng Chen, Xianwen Mao, invention disclosure "A competition-enabled imaging technique with super-resolution," filed on September 28, 2018; US patent filed September 27, 2019

PARTICIPANTS:

Participant Type: PD/PI

Participant: Peng Chen

Person Months Worked: 3.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

RPPR Final Report
as of 15-Oct-2021

Participant: Ming Zhao

Person Months Worked: 5.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Wenjie Li

Person Months Worked: 6.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Rong Ye

Person Months Worked: 15.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Xianwen Mao

Person Months Worked: 15.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Youngchan Park

Person Months Worked: 1.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Postdoctoral (scholar, fellow or other postdoctoral position)

Participant: Kushal Sengupta

Person Months Worked: 8.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Udit Chakraborty

Person Months Worked: 3.00

Project Contribution:

National Academy Member: N

Funding Support:

Participant Type: Graduate Student (research assistant)

Participant: Heting Pu

Person Months Worked: 12.00

Funding Support:

RPPR Final Report
as of 15-Oct-2021

Project Contribution:
National Academy Member: N

Participant Type: Research Experience for Undergraduates (REU) Participant

Participant: Zhaohong Wang

Person Months Worked: 2.00

Funding Support:

Project Contribution:

National Academy Member: N

Participant Type: Research Experience for Undergraduates (REU) Participant

Participant: Diego Garzon

Person Months Worked: 2.00

Funding Support:

Project Contribution:

National Academy Member: N

International Travel:

SWE	2 days
NLD	5 days
DEU	2 days
ESP	5 days
KOR	5 days
GBR	5 days
GBR	7 days
ITA	5 days
TWN	5 days
DEU	5 days
CAN	5 days
FRA	5 days
ESP	5 days
CHN	7 days
ISR	5 days
CHN	5 days
NLD	30 days

ARTICLES:

RPPR Final Report

as of 15-Oct-2021

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: ACS Nano

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acsnano.8b01338

Volume: 12 Issue: 6

First Page #: 5570

Date Submitted: 6/29/18 12:00AM

Date Published: 6/1/18 12:00AM

Publication Location:

Article Title: Imaging Catalytic Hotspots on Single Plasmonic Nanostructures via Correlated Super-Resolution and Electron Microscopy

Authors: Ningmu Zou, Guanqun Chen, Xianwen Mao, Hao Shen, Eric Choudhary, Xiaochun Zhou, Peng Chen

Keywords: hot electron mechanism; linked nanostructures; plasmonic catalytic hotspots; single-molecule catalysis imaging

Abstract: Surface-plasmon (SP) enhanced catalysis on plasmonic nanostructures brings opportunities to increase catalytic efficiency and alter catalytic selectivity. Understanding the underlying mechanism requires quantitative measurements of catalytic enhancement on these nanostructures, whose intrinsic structural heterogeneity presents experimental challenges. Using correlated super-resolution fluorescence microscopy and electron microscopy, here we report a quantitative visualization of SP-enhanced catalytic activity at the nanoscale within single plasmonic nanostructures. We focus on two Au- and Ag-based linked nanostructures that present plasmonic hotspots at nanoscale gaps. Spatially localized higher reaction rates at these gaps vs nongap regions report the SP-induced catalytic enhancements, which show direct correlations with the nanostructure geometries and local electric field enhancements. Furthermore, the catalytic enhancement scales quadratically with the local actual light intensity,

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nature Chemistry

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41557-018-0022-y

Volume: 10 Issue: 6

First Page #: 607

Date Submitted: 6/29/18 12:00AM

Date Published: 3/1/18 5:00AM

Publication Location:

Article Title: Cooperative communication within and between single nanocatalysts

Authors: Ningmu Zou, Xiaochun Zhou, Guanqun Chen, Nisha May Andoy, Won Jung, Guokun Liu, Peng Chen

Keywords: catalytic communication, single nanocatalysts, cooperativity, synergism

Abstract: Enzymes often show catalytic allostery in which reactions occurring at different sites communicate cooperatively over distances of up to a few nanometres. Whether such effects can occur with non-biological nanocatalysts remains unclear, even though these nanocatalysts can undergo restructuring and molecules can diffuse over catalyst surfaces. Here we report that phenomenologically similar, but mechanistically distinct, cooperative effects indeed exist for nanocatalysts. Using spatiotemporally resolved single-molecule catalysis imaging, we find that catalytic reactions on a single Pd or Au nanocatalyst can communicate with each other, probably via hopping of positively charged holes on the catalyst surface, over ~102 nanometres and with a temporal memory of ~101 to 102 seconds, giving rise to positive cooperativity among its surface active sites. Similar communication is also observed between individual nanocatalysts, however it operates via a molecular diffusion mechanism involving neg

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors
Acknowledged Federal Support: Y

RPPR Final Report as of 15-Oct-2021

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Journal of The Electrochemical Society

Publication Identifier Type: DOI

Publication Identifier: 10.1149/2.040195jes

Volume: 166

Issue: 5

First Page #: H3286

Date Submitted: 5/10/19 12:00AM

Date Published: 3/1/19 5:00AM

Publication Location:

Article Title: Correlated Single-Molecule Reaction Imaging and Photocurrent Measurements Reveal Underlying Rate Processes in Photoelectrochemical Water Splitting

Authors: Justin B. Sambur, Douglas P. Shepherd, Mahdi Hesari, Michael Van Erdewyk, Eric Choudhary, Peng C

Keywords: Single-molecule reaction imaging, photocatalysis, photocurrent,

Abstract: Understanding reaction processes at the semiconductor/electrolyte interface is critical to developing (photo)electrochemical energy conversion systems. For the water oxidation reaction at a photoanode in a solar water splitting cell, there are multiple reaction steps that produce surface adsorbed reaction intermediates, such as OH• and H₂O₂. Little is known about how the reaction intermediate surface concentrations evolve during the water oxidation reaction. Here we use single-molecule fluorescence microscopy to track the time-dependent surface concentration of Ti-OH• species on TiO₂ photoanodes under chopped light illumination. We correlate the reaction intermediate surface concentration with the net rate of photoelectrochemical water oxidation (i.e., the photocurrent). The reaction intermediate dynamics follow different time scales in its temporal evolution from the photocurrent dynamics. By fitting the temporal evolutions of Ti-OH• species and photocurrent over a range of light

...

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: ACS Catalysis

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acscatal.8b04926

Volume: 9

Issue: 3

First Page #: 1985

Date Submitted: 5/10/19 12:00AM

Date Published: 1/1/19 12:00AM

Publication Location:

Article Title: Analogy between Enzyme and Nanoparticle Catalysis: A Single-Molecule Perspective

Authors: Rong Ye, Xianwen Mao, Xiangcheng Sun, Peng Chen

Keywords: Single-molecule reaction imaging, single nanoparticles, single enzymes

Abstract: In this Viewpoint, we discuss the analogies between enzyme and NP catalysis based on selected recent studies of single-molecule enzyme and NP catalysis. Specifically, we focus on comparing reaction kinetics, static disorder, dynamic disorder, parallel reaction pathways, and allosteric effects between enzyme and NP catalysis at the single-molecule level. In the discussions of NPs, we mainly refer to metallic NPs, but we will cite selective references about other types of materials, such as semiconductors, layered double hydroxides, and zeolites, at the appropriate sections.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

RPPR Final Report as of 15-Oct-2021

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nano Letters

Publication Identifier Type: DOI

Publication Identifier: 10.1021/acs.nanolett.8b04188

Volume: 19

Issue: 2

First Page #: 958

Date Submitted: 5/10/19 12:00AM

Date Published: 1/1/19 12:00AM

Publication Location:

Article Title: Quantifying Photocurrent Loss of a Single Particle–Particle Interface in Nanostructured Photoelectrodes

Authors: Mahdi Hesari, Justin B. Sambur, Xianwen Mao, Won Jung, Peng Chen

Keywords: Subparticle photoelectrochemistry, interface losses, single nanorod photoelectrodes

Abstract: Particle–particle interfaces are ubiquitous in nanostructured photoelectrodes and photovoltaics, which are important devices for solar energy conversion. These interfaces are expected to cause performance losses in these devices, but how much loss they would incur is poorly defined. Here we use a subparticle photoelectrochemical current measurement, in combination with specific photoelectrode configurations, to quantify the current losses from single particle–particle interfaces formed between individual TiO₂ nanorods operating as photoanodes in aqueous electrolytes. We find that a single interface leads to ~20% photocurrent loss (i.e., ~80% retention of the original current). Such quantitative, first-of-its-kind, information provides a metric for guiding the optimization and design of nanostructured photoelectrodes and photovoltaics.

Distribution Statement: 3-Distribution authorized to U.S. Government Agencies and their contractors

Acknowledged Federal Support: Y

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published

Journal: Nature Chemistry

Publication Identifier Type: DOI

Publication Identifier: 10.1038/s41557-019-0288-8

Volume: 11

Issue:

First Page #: 687

Date Submitted: 8/13/19 12:00AM

Date Published:

Publication Location:

Article Title: Super-resolution imaging of nonfluorescent reactions via competition

Authors: Xianwen Mao, Chunming Liu, Mahdi Hesari, Ningmu Zou, Peng Chen

Keywords: Superresolution imaging, nonfluorescent reactions, photocatalysis

Abstract: Super-resolved fluorescence microscopy techniques have enabled substantial advances in the chemical and biological sciences. However, they can only interrogate entities that fluoresce, and most chemical or biological processes do not involve fluorescent species. Here we report a competition-enabled imaging technique with super-resolution (COMPEITS) that enables quantitative super-resolution imaging of non-fluorescent processes. It is based on the incorporation of competition into a single-molecule fluorescence-detection scheme. We demonstrate COMPEITS by investigating a photoelectrocatalytic reaction; we map, with nanometre precision, a non-fluorescent surface reaction that is important for water decontamination on single photocatalyst particles. The subparticle-level quantitative information of reactant adsorption affinities unambiguously decouples size- and shape-scaling laws on specific particle facets and uncovers a surprising biphasic shape dependence, leading to catalyst design p

Distribution Statement: 2-Distribution Limited to U.S. Government agencies only; report contains proprietary info

Acknowledged Federal Support: Y

RPPR Final Report

as of 15-Oct-2021

Publication Type: Journal Article Peer Reviewed: Y **Publication Status:** 1-Published
Journal: Physical Review Letters
Publication Identifier Type: DOI **Publication Identifier:** 10.1103/PhysRevLett.126.126001
Volume: 126 **Issue:** 12 **First Page #:** 126001
Date Submitted: 5/21/21 12:00AM **Date Published:** 3/1/21 5:00AM
Publication Location:

Article Title: Stochastic Kinetics of Nanocatalytic Systems

Authors: Jingyu Kang, Seong Jun Park, Ji-Hyun Kim, Peng Chen, Jaeyoung Sung

Keywords: nanoparticle catalysis, stochastic kinetics, theory

Abstract: Adsorption plays vital roles in many processes including catalysis, sensing, and nanomaterials design. However, quantifying molecular adsorption, especially at the nanoscale, is challenging, hindering the exploration of its utilization on nanomaterials that possess heterogeneity across different length scales. Here we map the adsorption of nonfluorescent small molecule/ion and polymer ligands on gold nanoparticles of various morphologies in situ under ambient solution conditions, in which these ligands are critical for the particles' physiochemical properties. We differentiate at nanometer resolution their adsorption affinities among different sites on the same nanoparticle and uncover positive/negative adsorption cooperativity, both essential for understanding adsorbate-surface interactions. Considering the surface density of adsorbed ligands, we further discover crossover behaviors of ligand adsorption between different particle facets, leading to a strategy and its implementation in

Distribution Statement: 1-Approved for public release; distribution is unlimited.

Acknowledged Federal Support: Y

Partners

I certify that the information in the report is complete and accurate:

Signature: Peng Chen

Signature Date: 8/6/21 11:16AM

Major Goals

The original 3-year grant and the 1-year add-on period comprise the following research goals:

- Develop novel methods to control plasmonic nanostructures and image reactions at high spatiotemporal resolution in operando, so as to quantify the kinetics and understand the mechanism of surface plasmon-enhanced catalysis on metal surfaces.
- Develop a method for super-resolution imaging of nonfluorescent reactions, named COMPEITS (competition-enabled imaging technique with superresolution).
- Continue the development of COMPEITS and its applications in quantifying/mapping molecular adsorption on catalyst particles.

Accomplished under Goals

Under the research goals, we have produced 9 publications (see list under “Dissemination,” and the *most significant results* are in the following publications as summarized below:

- Ningmu Zou,[†] Xiaochun Zhou,[†] Guanqun Chen,[†] Nesha May Andoy, Won Jung, Guokun Liu, Peng Chen* “Cooperative communication within and between single nanocatalysts” *Nature Chem.* **2018**, *10*, 607-614.

Summary: Enzymes often show catalytic allostery in which reactions occurring at different sites communicate cooperatively over distances of up to a few nanometers. Whether such effects can occur with non-biological nanocatalysts remains unclear, even though these nanocatalysts can undergo restructuring and molecules can diffuse over catalyst surfaces. Here we report that phenomenologically similar, but mechanistically distinct, cooperative effects indeed exist for nanocatalysts. Using spatiotemporally resolved single-molecule catalysis imaging, we find that catalytic reactions on a single Pd or Au nanocatalyst can communicate with each other, probably via hopping of positively charged holes on the catalyst surface, over $\sim 10^2$ nanometers and with a temporal memory of $\sim 10^1$ to 10^2 seconds, giving rise to positive cooperativity among its surface active sites. Similar communication is also observed between individual nanocatalysts, however it operates via a molecular diffusion mechanism involving negatively charged product molecules, and its communication distance is many micrometers. Generalization of these long-range intra- and interparticle catalytic communication mechanisms may introduce a novel conceptual framework for understanding nanoscale catalysis.

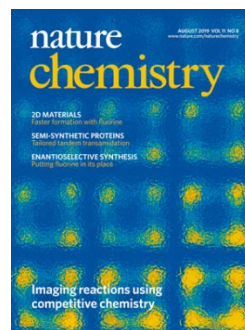
- Ningmu Zou, Guanqun Chen, Xianwen Mao, Hao Shen, Eric Choudhary, Xiaochun Zhou, Peng Chen* “Imaging catalytic hotspots on single plasmonic nanostructures via correlated super-resolution and electron microscopy” *ACS Nano* **2018**, *12*, 5570-5579.

Summary: Surface-plasmon (SP) enhanced catalysis on plasmonic nanostructures brings opportunities to increase catalytic efficiency and alter catalytic selectivity. Understanding the underlying mechanism requires quantitative measurements of catalytic enhancement on these nanostructures, whose intrinsic structural heterogeneity presents experimental challenges. Using correlated super-resolution fluorescence microscopy and electron microscopy, here we report a quantitative

visualization of SP-enhanced catalytic activity at the nanoscale within single plasmonic nanostructures. We focus on two Au- and Ag-based linked nanostructures that present plasmonic hotspots at nanoscale gaps. Spatially localized higher reaction rates at these gaps vs nongap regions report the SP-induced catalytic enhancements, which show direct correlations with the nanostructure geometries and local electric field enhancements. Furthermore, the catalytic enhancement scales quadratically with the local actual light intensity, attributable to hot electron involvement in the catalytic enhancement mechanism. These discoveries highlight the effectiveness of correlated super-resolution and electron microscopy in interrogating nanoscale catalytic properties.

- Xianwen Mao, Chunming Liu, Mahdi Hesari, Ningmu Zou, Peng Chen* “Super-resolution imaging of nonfluorescent reactions via competition” *Nature Chem.* **2019**, *11*, 687-694. Featured on journal cover.

Summary: Super-resolved fluorescence microscopy techniques have enabled substantial advances in the chemical and biological sciences. However, they can only interrogate entities that fluoresce, and most chemical or biological processes do not involve fluorescent species. Here we report a competition-enabled imaging technique with super-resolution (COMPEITS) that enables quantitative super-resolution imaging of non-fluorescent processes. It is based on the incorporation of competition into a single-molecule fluorescence-detection scheme. We demonstrate COMPEITS by investigating a photoelectrocatalytic reaction; we map, with nanometre precision, a non-fluorescent surface reaction that is important for water decontamination on single photocatalyst particles. The subparticle-level quantitative information of reactant adsorption affinities unambiguously decouples size- and shape-scaling laws on specific particle facets and uncovers a surprising biphasic shape dependence, leading to catalyst design principles for optimal reactant adsorption efficacy. With its ability to provide spatially resolved information on the behaviours of unlabelled, non-fluorescent entities under operando conditions, COMPEITS could interrogate a variety of surface processes in fields ranging from heterogeneous catalysis and materials engineering to nanotechnology and energy sciences.



- Rong Ye[†], Ming Zhao[†], Xianwen Mao, Zhaohong Wang, Diego A. Garzón, Heting Pu, Zhiheng Zhao, Peng Chen* “Nanoscale cooperative adsorption for materials control” *Nature Commun.* **2021**, *12*, 4287.

Summary: Adsorption plays vital roles in many processes including catalysis, sensing, and nanomaterials design. However, quantifying molecular adsorption, especially at the nanoscale, is challenging, hindering the exploration of its utilization on nanomaterials that possess heterogeneity across different length scales. Here we map the adsorption of nonfluorescent small molecule/ion and polymer ligands on gold nanoparticles of various morphologies in situ under ambient solution conditions, in which these ligands are critical for the particles’ physiochemical properties. We differentiate at nanometer resolution their adsorption affinities among different sites on the same nanoparticle and uncover positive/negative adsorption cooperativity, both essential for understanding adsorbate-surface interactions. Considering the surface density of adsorbed ligands, we further discover crossover behaviors of ligand adsorption between different particle facets, leading to a

strategy and its implementation in facet-controlled synthesis of colloidal metal nanoparticles by merely tuning the concentration of a single ligand.

Dissemination

During the entire funding period, we have had the following publications that were funded or partly funded by this award:

1. Ningmu Zou,[†] Xiaochun Zhou,[†] Guanqun Chen,[†] Nesha May Andoy, Won Jung, Guokun Liu, Peng Chen* “Cooperative communication within and between single nanocatalysts” *Nature Chem.* **2018**, *10*, 607-614.

News highlight:

- “Communicating catalysts,” *Nature Chem.* **2018**, *10*, 580-582.
 - “Nanoparticle catalysts communicate across distances,” *Chem. Eng. News* **2018**, *96*, 6.
 - “Understanding nanocatalysts' 'talk' could better inform design,” *Cornell Chronicle*, March 26, 2018.
 - “Communication is key in nanocatalysts too,” Nanotechweb.org, UK Institute of Physics.
2. Ningmu Zou, Guanqun Chen, Xianwen Mao, Hao Shen, Eric Choudhary, Xiaochun Zhou, Peng Chen* “Imaging catalytic hotspots on single plasmonic nanostructures via correlated super-resolution and electron microscopy” *ACS Nano* **2018**, *12*, 5570-5579.
 3. Rong Ye,[†] Xianwen Mao,[†] Xiangcheng Sun, Peng Chen* “Analogy between enzyme and nanoparticle catalysis: a single-molecule perspective” *ACS Catal.* **2019**, *9*, 1985-1992. Invited viewpoint.
 4. Mahdi Hesari, Justin Sambur, Xianwen Mao, Won Jung, Peng Chen* “Quantifying photocurrent loss of a single particle-particle interface in nanostructured photoelectrodes” *Nano Lett.* **2019**, *19*, 958-962.

News highlight:

- “A first: Cornell researchers quantify photocurrent loss in particle interface,” *Cornell Chronicle*, January 30, 2019
5. Justin B. Sambur,^{*} Douglas P. Shepherd, Mahdi Hesari, Michael Van Erdewyk, Eric Choudhary, and Peng Chen* “Correlated single-molecule reaction imaging and photocurrent measurements reveal underlying rate processes in photoelectrochemical water splitting” *J. Electrochem. Soc.* **2019**, *166*, H3286-H3293.
 6. Xianwen Mao, Chunming Liu, Mahdi Hesari, Ningmu Zou, Peng Chen* “Super-resolution imaging of nonfluorescent reactions via competition” *Nature Chem.* **2019**, *11*, 687-694. Featured on journal cover.

News highlight:

- “Extending fluorescence microscopy to nonfluorescent reactions,” *Chem. Eng. News*, July 12, 2019
 - “Breakthrough imaging of non-fluorescent reactions,” *Microscopy and Analysis* editorial, July 10, 2019
 - “New imaging method aids in water decontamination,” *Cornell Chronicle*, July 8, 2019
 - “Minder zien, meer weten,” Dutch news platform [C2w]
 - “Superresolution imaging method aids in water decontamination,” *Photonics* media
7. Ming Zhao, Peng Chen* “Exploring plasmonic photocatalysis via single-molecule reaction imaging” *Nano Lett.* **2020**, *20*, 2939-2940. Invited Perspective.

8. Jingyu Kang, Seong Jun Park, Peng Chen,* Jaeyoung Sung* “Stochastic kinetics of nanocatalytic systems” *Phys. Rev. Lett.* **2021**, *126*, 126001.
9. Rong Ye[†], Ming Zhao[†], Xianwen Mao, Zhaohong Wang, Diego A. Garzón, Heting Pu, Zhiheng Zhao, Peng Chen* “Nanoscale cooperative adsorption for materials control” *Nature Commun.* **2021**, *12*, 4287.

News highlight:

- Featured in *Nat Commun* Editors’ Highlights in Materials Science and Chemistry.
- "Small molecule plays outside role in controlling nanoparticle," *Cornell Chronicle*, July 13, 2021
- “Chemistry discovery could remove micropollutants from environment,” U.S. Army DEVCOM Army Research Laboratory Public Affairs; July 14, 2021

The PI has also given the following seminars:

1. Department of Physics, Chalmers University of Technology, Sweden, November 20, 2017
2. Nanoscale Photonic Imaging Collaborative Research Center, Georg August University, Göttingen, Germany, November 28, 2017
3. Institute for Molecules and Materials, Radboud University, the Netherlands, November 29, 2017
4. Department of Chemistry, University of Texas at Austin, February 21-22, 2018
5. Division of Chemistry and Chemical Engineering, CalTech, February 26-27, 2018
6. Center for Chemistry at the Space-Time Limit, UC Irvine, March 15, 2018
7. Department of Chemistry and Biochemistry, UC San Diego, March 16, 2018
8. Department of Chemistry and Biochemistry, University of Maryland, Baltimore County, April 13, 2018
9. Department of Chemistry, Seoul National University, South Korea, June 5, 2018
10. Department of Chemistry, Gwangju Institute of Science and Technology, South Korea, June 7, 2018
11. Sessler Distinguished Alumni Lecture, Department of Chemistry, Stanford University, November 8, 2018
12. Department of Chemistry and Biochemistry, Ohio State University, November 15, 2018
13. Department of Chemical Engineering, Virginia Tech, April 12, 2019
14. Department of Chemistry, Queens College, April 15, 2019
15. Center for Nanoscience and Nanotechnology, The Hebrew University of Jerusalem, September 8, 2019
16. Brian Bent Lecture, Department of Chemistry, Columbia University, December 5, 2019
17. Joan Van der Waals Lecture, Institute of Physics and Institute of Chemistry, University of Leiden, the Netherlands, December 12, 2019
18. Chemistry Virtual Seminar Series, Division of Chemistry and Biological Chemistry, Nanyang Technological University, Singapore, July 3, 2020
19. Modern Optics and Spectroscopy seminar series, MIT, September 29, 2020 (remote)
20. Center for Chemical Dynamics in Living Cells and the Department of Chemistry, Chung-Ang University, Seoul, South Korea; February 18, 2021 (remote)
21. Physical Chemistry seminar, University of Colorado, Boulder, March 19, 2021 (remote)

The PI has also given the following conference talks:

1. Symposium on “Nano and sub-nano clusters as the smallest and highly-tunable interfaces,” APS Meeting, Los Angeles, CA; March 5-9, 2018
2. Symposium on “Understanding the Complexity of the Nano-Bio Interface,” ACS National Meeting, New Orleans, LA; March 18-22, 2018
3. Plenary Speaker, 6th International Congress on Operando Spectroscopy, Estepona (Málaga, Spain); April 15-19, 2018
4. Plenary Speaker, Symposium of the Center for Chemical Dynamics in Living Cells, Chung-Ang University, Seoul, South Korea; June 4, 2018
5. Plenary Speaker, IBS Symposium on Nanomaterials and Spectroscopy, Korean Advanced Institute of Science and Technology, South Korea; June 8, 2018
6. Catalysis Workshop – East, Berkshires, MA; August 14-17, 2018
7. Symposium on "Technical Developments & Applications of Optical Chemical Imaging," ACS National Meeting, Boston, MA; August 19-23, 2018
8. Symposium on "Light-Nanomaterial Interactions for Ultrasensitive Electrochemical Sensing and Imaging and Materials Chemistry," ACS National Meeting, Boston, MA; August 19-23, 2018
9. Symposium on “Light and Electrochemistry in (Ordered) Nanostructures,” SciX 2018, annual meeting of Federation of Analytical Chemistry and Spectroscopy Societies (FACSS), Atlanta, GA; October 21-24, 2018
10. Symposium on “Emerging Materials and Characterization for Selective Catalysis,” Materials Research Society (MRS) Fall Meeting, Boston, MA; November 25-30, 2018
11. Keynote lecture, Workshop on probing chemical reactions by single-molecule spectroscopy, International Bunsen discussion meeting, Kloster Höchst, Germany; March 27-29, 2019
12. Symposium on “Emerging Frontiers in Fluorescence Microscopy, From Single Molecules to Super-Resolution” ACS National Meeting, Orlando, FL; March 31 – April 4, 2019
13. Chemical Pioneer Award address, annual meeting of The American Institute of Chemists Philadelphia, PA; May 9, 2019
14. Symposium on “Advances in Fluorescent Probe and Fluorescence Imaging Modalities,” Canadian Society of Chemistry, Quebec City, Canada; June 3-7, 2019
15. Symposium CT1: Advanced Methodologies for Matter Characterization and better Knowledge, 47th IUPAC World Chemistry Congress (WCC), Paris, France; July 7-12, 2019
16. Keynote lecture, 5th International Conference on Energy Conversion and Storage (5th ICECS), Peking University, Beijing, China; August 15-16, 2019
17. Keynote lecture, 16th Conference on Methods and Applications of Fluorescence (MAF 2019), UCSD; August 20-24, 2019
18. Catalysis: Small Molecules, Big Challenge, Technion, Israel; September 8-11, 2019
19. Keynote lecture, 18th Beijing Conference and Exhibition on Instrumental Analysis, Beijing, China; October 23-26, 2019
20. Thematic focus session on “Chemistry of Single Particles and Molecules: A Fluorescence Approach,” annual Dutch Chemistry Congress, CHAINS 2019; December 11, 2019
21. Symposium on "Spectroscopy for Understanding Catalysis," ACS National Meeting, San Francisco, CA; August 16-20, 2020
22. Keynote Lecture, symposium on “Cutting Edge Electrochemical Measurement Techniques,” 71st Annual Meeting of the International Society of Electrochemistry, Belgrade, Serbia; August 30 – September 4, 2020 (on-line)

23. Virtual Conference on Probing Chemical Reactions through Single-Molecule Spectroscopy; June 8, 2021

Honors

The PI has received the following honors during the funding period:

- 2020 Best Editor Award, *Nano Research*
2019 Chemical Pioneer Award
2018 Fellow, American Association for the Advancement of Science
2018 Bau Family Award in Inorganic Chemistry
2017 Visiting Professor in the Debye Chair, Utrecht University
- 2019 Joan Van der Waals Lecturer, University of Leiden
2019 Brian Bent Lecturer, Columbia University
2018 Sessler Distinguished Alumni Lecturer, Stanford University
- 2020 Keynote Lecture, 71st Annual Meeting of the International Society of Electrochemistry
2019 Keynote Lecture, 18th Beijing Conference and Exhibition on Instrumental Analysis
2019 Keynote Lecture, 16th Conference on Methods and Applications of Fluorescence (MAF 2019)
2019 Keynote Lecture, 5th International Conference on Energy Conversion and Storage (5th ICECS)
2019 Opening Keynote Lecture, International Bunsen Discussion Meeting on Probing Chemical Reactions by Single-molecule Spectroscopy
2018 Keynote Lecture, 14th European Biological Inorganic Chemistry Conference (EuroBIC-14)
2018 Plenary Lecture, IBS Symposium on Nanomaterials and Spectroscopy, Korean Advanced Institute of Science and Technology, South Korea
2018 Plenary Lecture, Symposium of the Center for Chemical Dynamics in Living Cells, Chung-Ang University, South Korea
2018 Plenary Lecture, 6th International Congress on Operando Spectroscopy