Yoshiyasu Matsumoto
Emeritus Professor, Kyoto University

[Present Position]
Fellow at Toyota Physical and Chemical Research Institute

[Research Field]
Physical Chemistry, Spectroscopy and Dynamics, Catalysts, Materials Science

[Date of Birth]
January 20, 1953 **[Sex]** Male **[Nationality]** Japan

[Graduation and Degree]
3/1975 Faculty of Engineering, Department of Industrial Chemistry. Kyoto University
3/1977 Graduate School of Engineering, Department of Industrial Chemistry, Master Course, Kyoto University
6/1981 Graduate School of Engineering, Department of Reaction Chemistry, Doctor Course, The University of Tokyo
6/1981 Doctor of Engineering, The University of Tokyo (Thesis title: A Study on Gas Phase Reactions by CARS)

[Professional Career]

NSF postdoctoral fellow, Department of Chemistry, University of Pittsburgh	7/1981-8/1984
Research Assistant, Institute for Molecular Science	9/1984-1/1985
Research Scientist, RIKEN (Institute of Physical and Chemical Research)	2/1985-2/1990
Associate Professor, Institute for Molecular Science	3/1990-3/1997
Professor, The Graduate University for Advanced Studies (Sokendai)	4/1997- 3/2004
Chair, Department of Photoscience, The Graduate University for Advanced Studies (Sokendai)	4/1999- 3/2001
Dean, School for Advanced Sciences, The Graduate University for Advanced Studies (Sokendai)	4/2001- 3/2004
Professor, Institute for Molecular Science	4/2004-3/2007
Professor, Kyoto University, Graduate School of Science	4/2007- 3/2018
Chair of Department of Chemistry, Kyoto University	4/2010- 3/2012
Vice Dean of Graduate School of Science, Kyoto University	4/2016- 3/2018
Fellow, Toyota Institute of Physical and Chemical Research	4/2018 – present

[Membership]
Chemical Society of Japan, Physical Society of Japan, Japan Society for Molecular Science, The Surface Science Society of Japan, The Japanese Photochemistry Association

[Honors and Awards]

Hanse Wissenschaftskolleg (Fellow of Hanse Institute for Advanced Studies), Germany	8/2002
The CSJ(Chemical Society Japan) Award for Creative Work	3/2006

Summary of research by Dr. Yoshiyasu Matsumoto

The research of Dr. Y. Matsumoto has been focused on a wide range of subjects, from the most fundamental ones, such as adsorbates on well-defined single crystal surfaces, to complex systems of importance in applications such as photocatalyst particles. He has promoted these research works, always aiming to understand them at the molecular level. His major research topics are: (1) the structure and dynamics of van der Waals molecular complex, (2) the electron excited state dynamics and charge transport in organic molecules, (3) the photochemistry and photoinduced ultrafast dynamics at metal surfaces, (4) the structure and molecular orientation in and at the surface/interface of ice thin film crystals grown on metal, and (5) the charge dynamics in photocatalyst particles. The following are the main results of these topics.

1. Structure and dynamics of van der Waals molecular complex

Molecular complexes formed by weak intermolecular forces such as van der Waals forces have vibration modes with large amplitudes that are not found in ordinary stable molecules. Thus, exploring the structure and dynamics of weakly bound complexes is useful for promoting an understanding of intermolecular interactions. He prepared complexes of BF_3 and rare gas atoms, acetylene dimer, NO dimer etc using a supersonic jet, and performed high resolution infrared absorption spectroscopy of these molecular complexes. Although acetylene dimer has a T-shaped stable structure, he demonstrated for the first time that two acetylene molecules undergo large-amplitude motion in which molecules rotate like a gear. Moreover, he found that the line width of the asymmetric stretching vibration band of NO dimer greatly differs from that of the symmetrical stretching vibration of NO, showing that the vibrational predissociation rate of the complex strongly depends on vibrational modes.

2. Electronically excited state dynamics and charge transport in organic molecules

The dynamics of electronic states of organic molecules is an central issue that has been studied for a long time. Much effort has be devoted to understand the dynamics of excitons in crystals and thin films composed of organic molecules, the transport of charges derived from charge separation of excitons in organic molecule aggregates. Moreover, this research area is very important in applications such as photovoltaics. He expanded the research targets from isolated molecules in the gas phase to organic

semiconductor molecular aggregates in the forms of crystals and thin films, and elucidated the electronic excited state dynamics and charge transfer / transport of organic molecules.

2-1. Intramolecular electron relaxation dynamics in isolated vapor phase molecules

An excited singlet state relaxes to an excited triplet state due to spin-orbit interaction: intersystem crossing. This process is a research subject that has been of great scientific interest for years. However, in the conventional studies targeting molecules in the gas phase, intermolecular collisions cannot be avoided and it is impossible to cool down to extremely low temperature without molecular aggregation. Thus, organic molecules are distributed among many ro-vibrational states of the electronic ground state. This makes difficult to investigate the molecular rotation state dependence of intramolecular electron relaxation dynamics of polyatomic molecules as large as benzene. He simplified the spectrum of organic molecules by lowering the internal temperature of molecules by using supersonic jet expansion, and clarified how the spin-orbit interaction depends molecular rotation in the excited state using an excitation laser with a linewidth sufficiently narrow to resolve rotational structures in excitation spectra. The molecules of interest are pyrazine, pyrimidine and the like. He succeeded in coherently exciting molecular eigenstates in the singlet manifold that are hybridized with triplet ro-vibrational states with spin-orbit coupling, and clarified that the number of triplet states involved in this hybridization depends on the rotational quantum number of states in the singlet manifold.

2-2. Exciton dynamics in organic semiconductor thin films

The properties of excitons in organic semiconductors are important for determining the light conversion efficiency in photovoltaics. Because molecules in organic semiconductor crystals are bound with weak intermolecular forces, the spatial extent, lifetime, diffusion and other properties of excitons are very much different from those of inorganic semiconductors with strong bonds between constituent atoms. He focused on singlet fission: a singlet exciton excited by one photon splits into two triplet excitons. This is an important process capable of doubling the light conversion efficiency of organic semiconductor solar cells. Focusing on rubrene single crystal, he demonstrated that single fission takes place from superposition states of excited singlet and triplet states coherently excited and it is promoted by symmetry breaking with excitation of intermolecular vibrational modes along with the electronic transition. This indicates that coherent excitation of the superposition states in the electronically

excited states is also important in aggregates just as in the case of isolated molecules in the gas phase. It is also the first example that clearly demonstrates experimentally that singlet fission is promoted through vibronic interaction associated with intermolecular vibration.

2-3. Charge transport in organic semiconductor thin film

Charge transport in organic semiconductor thin films is an important process determining the performance of organic EL elements, organic field effect transistors, and the like. He succeeded for the first time in mapping the two dimensional spatial distribution of the carrier density in the organic field effect transistor made of pentacene by a home-built sum frequency generation spectroscopy microscope. Furthermore, he succeeded in observing the absorption spectra of holes responsible for charge transport by charge modulation spectroscopy for an organic field effect transistor composed of rubrene, and also succeeded in clarifying that charge transport in dinaphtho [2,3-b: 2',3'-f] thieno [3,2-b] thiophene (DNTT) is greatly enhanced by introducing side chain of alkyl group into this molecule.

3. Photochemistry and Photoinduced Ultrafast Dynamics of Adsorbed Species on Metal Surface

3-1. Photochemistry on metal solid surface

Two excitation mechanisms can be considered for the excitation of electronic states of adsorbed species on the metal surface by light irradiation: light absorption (direct excitation) of the adsorbed species itself and light absorption (indirect excitation) by the metal surface. It is difficult to distinguish these with experiments, and it has been considered that nearly all photochemistry in the visible and ultraviolet regions on metal surfaces is induced by indirect excitation. He studied the photochemistry of methane weakly adsorbed on a clean metal surface. It was known that methane is dissociated in the gas phase by light absorption in the vacuum ultraviolet region. However, he discovered for the first time that methane physisorbed on platinum, palladium, copper, etc. dissociates into methyl and hydrogen by ultraviolet light irradiation at the wavelength where methane in the gas phase has no absorption. Furthermore, he made a systematic measurement of the photochemical cross section and clarified that the excitation mechanism is a transition from the highest occupied state of methane: the direct excitation of the methane electronic state. In addition to this, he also studied the surface reaction of highly energetic oxygen atoms generated by photodissociation of N_2O on Pt(111), and put forward the hot atom surface chemistry.

3-2. Photo-induced ultrafast dynamics of adsorbed species on metal surfaces

He conducted research on real-time observation of ultrafast dynamics of surface photochemical reaction process. When an adsorbed molecule is electronically excited by light, vibration of adsorbed species is excited through electron-phonon interaction, but nuclear dynamics at metal surfaces induced by the electronic excitation was rarely explored. He applied femtosecond time-resolved surface second harmonic generation spectroscopy to alkali metal atoms adsorbed on platinum or copper surfaces. He observed that the stretching vibration between alkali atom and substrate is excited in phase: coherent phonon excitation. In addition, he clarified the mechanism of relaxation dynamics of this coherent vibration, and succeeded for the first time in exciting the surface vibration mode selectively by shaping the sequence of excitation light pulses. He also showed that direct and indirect excitations selectively occur depending on excitation wavelength. Thermal reactions on slid surface take place during excitation of various vibration modes of adsorption systems under the thermal equilibrium, in which some vibration modes are directly involved in the reaction. Thus, the mode-selective surface vibration excitation by light-pulse irradiation opens a way for controlling surface reaction. Furthermore, he expanded the range of research into the photo-stimulated desorption dynamics of CO, which is a simplest chemisorbed polyatomic molecule, and newly applied time-resolved vibrational spectroscopy, infrared and visible sum frequency generation spectroscopy, to this adsorption system. As a result, he succeeded in finely grasping the adsorbate nuclear motions along the dissociation of the bond between the adsorbed species and the surface: desorption of CO molecule to the gas phase. This research can be regarded as a breakthrough in elucidating the surface ultrafast process of more complex and generalized polyatomic molecular adsorption species.

4. Molecular arrangement and structure on ice thin film crystal and its surface

Since the study of the structure of ice crystals by X-ray diffraction in the 1930's, the ice exhibiting ferroelectricity in which water molecules orient uniformly has gathered a lot of attentions. He elucidated how the ferroelectric ice thin film grows by controlling the orientation of water molecules by sum frequency generation spectroscopy. The growth of a ferroelectric ice thin-film crystal on Pt (111) was a long standing controversial issue. He has put an end to this controversy by showing that the crystal thin film in which water molecules are oriented in the same direction on Pt(111). Also noteworthy is that heterodyne detection of sum frequency generation signals allows to determine the orientation of water molecules in the ice thin crystal: water molecules at

the Pt(111) surface adsorb while one of hydrogen atoms points to the surface, so that this molecular orientation is propagated into the thin film through hydrogen bonds. Furthermore, he elucidated the details of ice structure at the vacuum side of the ice thin film crystals, where the outermost surface and subsurface has a structure different from that in the bulk thin film.

5. Charge dynamics in photocatalyst particles

He elucidated the dynamics of photoinduced carriers of inorganic semiconductor particles. One of the most important applications of inorganic semiconductors is photocatalytic reactions. Water decomposition by sunlight using photocatalyst is a very important research subject, because it can supply sustainable energy with hydrogen as an energy source to the world: a hydrogen-based society. The light conversion efficiency in photocatalytic water decomposition using inorganic semiconductor particles has been still low, and researches aiming to improve the efficiency have been actively made. This low light conversion efficiency is attributable to the fact that charge recombination in the grain or the grain boundary is extremely fast as compared with oxidation/reduction reaction rates. Moreover, in conventional research methods, samples with various sizes, shapes and aggregation degrees are measured by dispersing them in water. Thus, it is difficult to disentangle many kinds of elementary processes involved in the photocatalytic reactions at the molecular level. He has demonstrated that the carrier recombination rate of catalysts developed for practical use is lowered by depositing cocatalysts. Also, unlike conventional photocatalyst studies, he first discovered that water adsorption improves the hole trapping ability on the catalyst surface by controlling the number of quasi-liquid layers on the catalyst particle surface.

Publication list (2018)

Yoshiyasu Matsumoto

Original Papers

1. "Molar excess enthalpies for water + ethanediol, +1,2-propanediole and + 1,3-propanediole at 298.15K",
   Y. Matsumoto, N, Touhara, N. Nakanishi, and N. Watanabe,
   Journal of Chemical Thermodynamics **9**, 801 (1977).

2. "Thermal decomposition of ammonia in shock waves",
   M. Yumura, A. Asaba, Y. Matsumoto, and H. Matsui,
   International Journal of Chemical Kinetics **12**, 439 (1980).

3. "Measurements of CARS intensity in hydrogen molecule behind shock waves",
   Y. Matsumoto, H. Matsui, and T. Asaba,
   Transactions of Japan Society for Aeronautical and Space Sciences **26**, 131 (1983).

4. "Direct excitation of triplet states in supersonic jets. Rotationally resolved ^3A_u-^1A_g laser induced phosphorescence",
   L.H. Spangler, Y. Matsumoto and D.W. Pratt,
   Journal of Physical Chemistry **87**, 4781 (1983).

5. "On the origin of the rotational state dependence of the decay of intermediate case molecules. Role of angular momentum selection rules in intersystem crossing",
   Y. Matsumoto, L.H. Spangler, and D.W. Pratt,
   Chemical Physics Letters **98**, 333 (1983).

6. "Time-resolved fluorescence depolarization in the decay of intermediate case molecules. Zero-field level crossing of the molecular eigenstates of $^1B_{3u}$ pyrazine",
   Y. Matsumoto, L.H. Spangler, and D.W. Pratt,
   Chemical Physics Letters **95**, 343 (1983).

7. "$^1B_{3u}$ Pyrazine. Experimental tests of the theory of radiationless transitions",
   Y. Matsumoto, L.H. Spangler, and D.W. Pratt,
   Laser Chemistry **2**, 91 (1983).

8. "Singlet-triplet perturbations in pyrimidine. Magnetic field effects on collision-induced intersystem crossing",
   Y. Matsumoto and D.W. Pratt,
   Journal of Chemical Physics **81**, 573 (1984).

9. "Intersystem crossing in isolated molecules. Magnetic field effects on the fluorescence decay behavior of $^1B_{3u}$ pyrazine with single rovibronic level excitation",
   Y. Matsumoto, L.H. Spangler, and D.W. Pratt,
   Journal of Chemical Physics **80**, 5539 (1984).

10. "Pure rotational spectrum of SnH_4 in the vibrational ground state observed by infrared-radio frequency double resonance",

Y. Ohshima, Y. Matsumoto, M. Takami, and K. Kuchitsu,
Journal of Chemical Physics **85**, 5519 (1986).

11. "Free jet infrared absorption spectroscopy of the ν_3 band of TeF_6",
Y. Matsumoto and M. Takami,
Journal of Chemical Physics **85**, 3785 (1986).

12. "High-resolution infrared absorption spectroscopy of the CF_3I ν_2 band",
Y. Matsumoto, M. Takami, and P.A. Hackett,
Journal of Molecular Spectroscopy **118**, 310 (1986).

13. "Free jet infrared absorption spectroscopy of the CO stretching ν_6 fundamental of $Fe(CO)_5$",
Y. Matsumoto, T. Majima, and M. Takami,
Molecular Physics **61**, 1045 (1987).

14. "Infrared-microwave double resonance and diode laser spectroscopy of the ν_2/ν_4 bands of SnH_4",
Y. Ohshima, Y. Matsumoto, M. Takami, S. Yamamoto, and K. Kuchitsu,
Journal of Chemical Physics **87**, 5141 (1987).

15. "Free jet infrared absorption spectroscopy of stable and unstable molecular species",
M. Takami, Y. Ohshima, S. Yamamoto, and Y. Matsumoto,
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16. "Determination of the centrifugal-distortion-induced dipole moment of SnH_4 by infrared double resonance Stark spectroscopy",
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Journal of Chemical Physics **88**, 6747 (1988).

17. "Free-jet infrared absorption spectroscopy of the $(N_2O)_2$ van der Waals complex in the 8 μm region",
Y. Ohshima, Y. Matsumoto, M. Takami, and K. Kuchitsu,
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Y. Ohshima, Y. Matsumoto, M. Takami, and K. Kuchitsu,
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21. "Free jet infrared absorption spectroscopy of the ν_3 band of MoF_6",
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Molecular Physics **64**, 645 (1988).

22. "Ultra high-resolution fluorescence excitation spectrum of $^1B_1$ pyrimidine in a molecular beam. Structural assignments, analysis of singlet-triplet perturbations, and implications for intersystem crossing in the isolated molecule",
J.A. Konings, W.A. Majewski, Y. Matsumoto, D.W. Pratt, and W.L. Meerts,
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T. Majima, T. Ishii, Y. Matsumoto and M. Takami,
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24. "SF_6-sensitized infrared photodecomposition of $Fe(CO)_5$",
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25. "Highly excited even Rydberg series of Lu I studied by two-step laser photoionization spectroscopy",
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26. "Free-jet infrared spectroscopy of rare gas-$^{11}BF_3$ complexes in the 7 μm region",
Y. Matsumoto, Y. Ohshima, M. Takami, and K. Kuchitsu,
Journal of Chemical Physics **90**, 7017 (1989).

27. "Infrared-microwave double resonance and diode laser spectroscopy of the ν_1/ν_3 dyad of SnH_4",
L. Jörissen, Y. Ohshima, Y. Matsumoto, M. Takami, and K. Kuchitsu,
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28. "Mode-specific infrared photodissociation of nitric oxide dimers: High-resolution infrared spectroscopy of $(^{14}NO)_2$ and $(^{15}NO)_2$",
Y. Matsumoto, Y. Ohshima, and M. Takami,
Journal of Chemical Physics **92**, 937 (1990).

29. "Anomalous band shifts in the 14 μm infrared absorption spectra of rare-gas-BF_3 complexes",
G.-H Lee, Y. Matsuo, M. Takami, and Y. Matsumoto,
Journal of Chemical Physics **96**, 4079 (1992).

30. "Structure of adsorbed N_2O on a Pt(111) surface and photodissociation at 193 nm: effective formation of oxygen adatoms",
K.Sawabe and Y.Matsumoto,
Chemical Physics Letters **194**, 45 (1992).

31. "On the SF_6-sensitized IR photodecomposition of $Fe(CO)_5$: 5μm transient absorption measurements and absorption energy measurements",
T.Majima, Y.Matsumoto, and M.Takami,
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32. "Free-jet infrared absorption spectroscopy of the C_2H_2-Ar complex in the doubly degenerate monomer C-H bending region",
Y. Ohshima, Y. Matsumoto, M. Takami, and K. Kuchitsu,
Journal of Chemical Physics **99**, 8385 (1993).

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K. Sawabe, J. Lee, and Y. Matsumoto,
Journal of Chemical Physics **99**, 3143 (1993).

34. "Laser-induced photochemistry of nitrous oxide on a Pt(111) surface",
K. Sawabe and Y. Matsumoto,
Surface Science **283**, 126 (1993).

35. "Photochemistry and photodissociation dynamics of N_2O on metal surfaces",
Y. Matsumoto and K. Sawabe and J. Lee,
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36. "Photochemical C-H bond activation of methane on a Pt(111) surface",
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37. "Oxygen-exchange reaction between O_2 and NO coadsorbed on a Pt(111) surface: Reactivity of molecularly adsorbed oxygen",
K. Sawabe and Y. Matsumoto,
Surface Science **303**, L385 (1994).

38. "Dynamics of photochemical processes of N_2O adsorbed on metal and semiconductor surfaces",
Y. Matsumoto and J. Lee and H. Kato and K. Sawabe
Proceedings of SPIE - The International Society for Optical Engineering **2125**, 303 (1994).

39. "Free jet IR spectroscopy of SiF_4-rare gas complexes",
R.-D.Urban, L.G.Jörissen, Y.Matsumoto, and M.Takami,
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40. "The reactivity of molecular and atomic oxygen in oxygen-exchange reaction between NO and O_2 coadsorbed on a Pt(111) surface",
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