Academic background

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Work history

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1. Introduction to Research Topic at Toyota Physical and Chemical Research Institute

Research Topic; *Basic Studies and Applications of Far-ultraviolet spectroscopy*

"**Research purpose**" We have established attenuated total reflection (ATR)-far-ultraviolet (FUV) spectroscopy. It has enabled to explore electronic states and transitions of almost all kinds of molecules in condensed phase. Our previous research achievements can be divided into as follows; (i) Studies of electronic transitions and states including Rydberg (Ryd) states of organic molecules and polymers. (ii) Applications to materials sciences (polymers, carbon nanomaterials, ionic liquids, etc.). (iii) Investigations on water, aqueous solutions, and surface adsorption water. (iv) Electronic states and photocatalytic activities of TiO_2 and metal (Pt, Pd, and Au)-modified TiO_2 powders. (v) Proposal of FUV-DUV surface plasmon resonance (SPR) sensors using Al thin films.

At Toyota Physical and Chemical Research Institute I plan to expand the research topic (i). As for (i) we carried out detailed research on electronic transitions and states of alkanes, alcohols, amides, Nylon 6, graphenes and so on using spectral measurements and quantum chemical calculations. I will continue similar research for benzene, cyclohexane, and carbon nanotubes with Prof. Yusuke Morisawa (Kinki University). We will investigate not only electronic transitions and states but also stable structures, inter- and intramolecular interactions and the effects of molecular interactions on valence electrons (σ electrons) of single bonds of a molecule.

"**Methods**" We start from the ATR-FUV spectral measurements and quantum chemical calculations of cyclohexane, methyl- and dimethyl cyclohexane to investigate their changes in electronic transitions, electronic states, the most stable structures and energy levels by the differences in the number and positions of substituents and dihedral angle. We proceed the research as follows; (i) Comparison of the FUV spectra of cyclohexane, methyl- and dimethyl cyclohexane with those of *n*-alkanes and branched alkanes. (ii) Comparison of FUV spectra of dimethyl cyclohexane with different positions (o-, m-, p-) and different dihedral angles (cis-, trans-). (iii) Optimization of structure and calculations of electronic structure and transitions using Gaussian 09. (iv) A study of phase transition of cyclohexane by its measurements of temperature-dependent FUV spectral variations. We will elucidate how energy, stable structure and electronic structure change with the position of substituents and dihedral angle. Moreover, we will provide deeper insight into electronic translons in the FUV regions based on our new research including studies of benzene and carbon nanotubes together with our previous studies.

"Expected outcome" We may be able to obtain new insight into electronic states, electronic transitions, the most stable structures and inter- and intra-molecular interactions of cycloalkanes and benzene. For example, as for a cyclohexane ring it has been well known that a six-membered ring having a substitute at axial position is more stable than that at equatorial position. Our study may give convincing evidence for that. It is also possible for our study to yield new knowledge about effects of intermolecular interactions on valence electrons (σ electrons) of single bonds of a molecule from the temperature dependent FUV measurements. It is only our study of tetradecane that a change in the transition of an alkane by a change in σ orbital has ever been observed. The proposed investigation may pave the way to new σ chemistry.

2. Previous Research Achievements

Research Title; *Creation of Molecular Spectroscopy of Electronic and Vibrational Transitions in Condensed Phase and Its Application to Chemistry and Biomedicine*

I have aimed at establishing a world COE for molecular spectroscopy. I have long been involved in molecular spectroscopy research from far-ultraviolet (FUV) to far-infrared (FIR)/terahertz and Raman spectroscopy (1). One of the strategies of my spectroscopy research is to challenge undeveloped and unexplored research fields such as FUV spectroscopy of condensed matters (1). Another important strategy has been to carry out comprehensive investigations of molecular spectroscopy, from investigations of principles (for example, studies of the mechanism of surface-enhanced Raman scattering), developments of cutting-edge instruments (for example, attenuated total reflection (ATR) FUV spectrometers), proposals of novel spectral analysis methods, and opening up new application fields (1). I have developed molecular spectroscopy markedly by my comprehensive and pioneering investigations in molecular spectroscopy (vibrational spectroscopy and electronic spectroscopy) in condensed phase. I have made extensive achievements in a wide range of spectroscopy covering FUV, UV, near-infrared (NIR), infrared (IR), FIR/terahertz (THz), and Raman spectroscopy. Among my path breaking achievements, the following six topics are chosen here, and their scientific significance is described.

1. Our Molecular Spectroscopy Research Common in the Whole Spectral Region from FUV to FIR/THz and Raman Spectroscopy

Usually, it is important to study molecular spectroscopy by dividing it into each spectroscopy such as IR, NIR, and FIR spectroscopy. However, at the same time, it is also profound to explore a wide range of spectral region comprehensively (1). As comprehensive research of molecular spectroscopy, we carried out (1); 1) application of quantum chemistry to molecular spectroscopy, 2) development of spectral analysis methods useful for various fields of molecular spectroscopy, 3) comparison of spectra between different spectroscopies for band assignments such as comparison between NIR and IR spectra.

In the applications of quantum chemistry, we have aimed at **making a strong bridge between molecular spectroscopy and quantum chemistry.** (2) Quantum chemical calculations have been extensively used in IR and Raman spectroscopy. Our group has expanded them to FUV, NIR and FIR/THz regions both to electronic spectra and vibrational spectra to reproduce their spectra and band assignments (1-8). Our significant achievements in the applications of quantum chemistry are as follows: 1) We introduced anharmonic calculations into NIR region (4,5). Using anharmonic calculations we have succeeded in reproducing many NIR spectra including those of alcohols (Figure 1), long chain fatty acids, nucleic acid bases, and natural products.

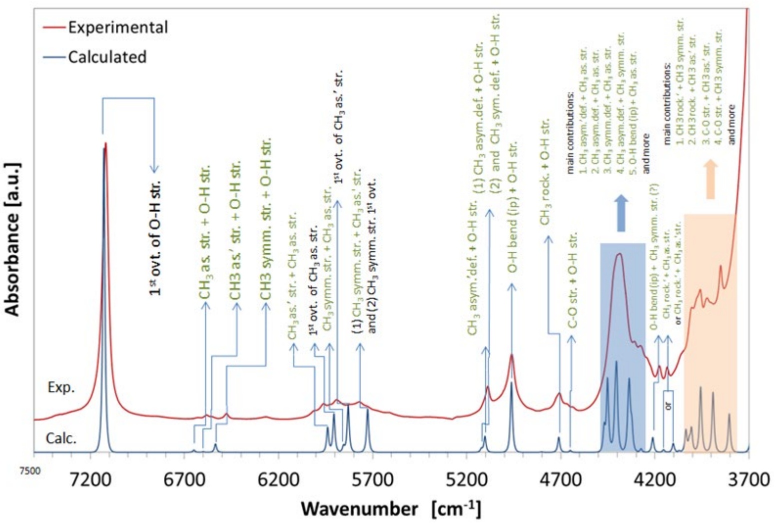


Figure 1. Examination of the NIR spectrum of low concentration (0.005 M) methanol by the calculated NIR spectrum. K.B.Bec, Y. Futami, M.J. Wojcik, Y. Ozaki, *Phys. Chem. Chem. Phys.* 18,13666 (2016).

Using quantum chemical calculations and NIR spectra measurements we also explored vibrational potential, anharmonicity, and molecular structures, particularly hydrogen bonding and intermolecular interactions (4,5). 2) In the studies of FUV spectroscopy we applied symmetry adapted cluster/configuration interaction (SAC-CI) method to develop investigations of electronic structures and transitions including Rydberg states (8). The quantum chemical calculation study also allowed Morisawa et al. to explore intermolecular interactions between alkanes in a crystal (9a). 3) We adopted Cartesian coordinate tensor transfer (CCT) method to disclose the band assignments in the FIR/THz spectra of polymers (6,7). This method enabled us to consider explicitly intermolecular interactions among polymer chains. By this method, we could not only make band assignments but also investigate intermolecular interactions and thermal expansion of polymers.

As for the second research topic “development of spectral analysis methods useful for various fields of molecular spectroscopy”, we have prominent achievements in chemometrics and two-dimensional correlation spectroscopy (2D COS). In the former, we developed sample and wavelength selection methods to build calibration curves with high accuracy efficiently. Among the chemometrics algorithms which we proposed Moving Window Partial Least Squares Regression (MWPLSR) (10). It has been used extensively as a powerful wavelength selection method for IR, Raman and NIR spectra.

For the studies on “comparison of spectra between different spectroscopies for band assignments”, besides conventional methods we have utilized 2D COS, which allows the direct comparison between two different spectroscopies by disclosing 2D correlation between them.

2. Development of attenuated total reflection (ATR)-FUV spectroscopy - Pioneering of novel σ chemistry.

In the FUV region (145-200 nm) a number of electronic transitions are expected to appear, however, absorption spectroscopy in the FUV region of condensed matters was an unexplored area of molecular spectroscopy (8). We developed a novel FUV spectrometer (145-300 nm) based on an ATR technique, making possible the easy measurement of FUV spectra of molecules in condensed phase (8,9,11). Since in the FUV region one can expect transitions due to π electrons, lone pair electrons, and σ electrons, the ATR-FUV method has allowed to investigate electronic states of almost all kinds of molecules in condensed phase. We established an assignment method of FUV spectra based on comparison of an observed FUV spectrum in condensed phase with the corresponding spectrum in gas phase and electronic transitions calculated by quantum chemical calculations (8,9). For example, in the study of electronically excited states of alkanes we assigned a band near 153 nm to a transition from HOMO-2 or HOMO-1 to 3*p* Rydberg state (Figure 2)(9b). Since alkanes are formed only by single bonds and do not have lone pair electrons, this transition reflects the state of σ electrons of alkanes. We measured ATR-FUV spectra of alkanes, alcohols, ketones, amides, benzene,,,, and so on and calculated their electronic transitions and electronic structure (9). We obtained a number of interesting results not only on electronic states and transitions where Rydberg orbitals are involved, but also on the effects of molecular interactions on valence electrons (σ electrons) of single bonds of a molecule.

FUV spectroscopy allowed Morisawa et al. (9a) to explore interactions between alkanes in a crystal, which are very important to the understanding of interactions that involve hydrogen atoms with same polarity such as CH…HC. We measured temperature-dependent ATR–FUV spectra of *n*-tetradecane during cooling and heating between 15 to −38 °C (9a). It was found that the band at 153 nm in the liquid phase becomes weaker, and instead, new bands appear at around 200 and 230 nm with decreasing temperature. Of note is that the conversion from the 153 nm band to the 200 and 230 nm bands occurs at the melting temperature. Therefore, it is very likely that the changes in the electronic states are induced by changes during the phase transition.

Figure 2. Calculated Kohn-Sham orbitals relevant for the excitation of *n*-hexane. (ref.9b).

This study suggests that an unusually compressed structure is generated on the surface of *n*-tetradecane at low temperatures and that this reversible phase change is responsible for the unusual absorption changes observed in the ATR–FUV spectra (9a). The ATR-FUV spectra of the low-temperature solid *n*-alkane indicate that the energy of its HOMO and the energy gap between its HOMO and LUMO in the solid phase are reduced to approximately 60% of those in the liquid phase. It is likely that σ electrons are affected by an intermolecular interaction in condensed phase.

We demonstrated a number of important applications of FUV spectroscopy (8).

i) One can apply FUV spectroscopy to qualitative analysis and discrimination analysis of various kinds of liquid and solid samples because each compound shows a characteristic FUV spectrum.
ii) We reveled that ATR-FUV spectroscopy is powerful for water research, from pure water, aqueous solutions, spring water, commercial mineral water, to surface adsorption water. Goto et al. explored structure of liquid water on an aluminum surface by variable-angle (VA) ATR-FUV spectroscopy (12).
iii) FUV spectroscopy has provided new opportunities for materials research (3b,13,14). It has been applied to studies of polymers, inorganic semiconductor materials, ionic liquids, and nanomaterials. Tanabe et al. applied the ATR-FUV-DUV spectroscopy to investigate electronic states and photocatalytic activities of TiO_2 and metal (Pt, Pd, and Au)-modified TiO_2 powders (13). Recently, Tanabe et al. used ATR-FUV/DUV (deep-ultraviolet) spectroscopy for electrochemistry research.
iv) We developed a nanosecond pump-probe transient FUV spectrometer which can be used to probe radical species in chain reactions such as O_3 pulse-photolytic reaction in an aqueous solution.
v) Tanabe et al. proposed FUV-DUV surface plasmon resonance (SPR) sensors using Al thin films (15). The SPR properties of the Al thin films with varying reflective index have been investigated by ATR-FUV spectrometer.

In this way we have demonstrated the great potential of FUV spectroscopy for condensed matter in both basic science and applications (8).

3. Establishment of near-infrared spectroscopy as a molecular spectroscopy.

Our contributions to NIR spectroscopy cover almost its whole range from basic studies of NIR spectroscopy such as investigations of overtones, combinations, vibrational potentials, and anharmonicities, developments of NIR spectrometers (e.g. surface plasmon resonance (SPR)-NIR spectroscopy, portable NIR imaging systems), experimental techniques, and spectral analysis methods, to a variety of applications such as applications to physical chemistry (water structure, hydration, hydrogen bindings, solution chemistry), biomedical applications, process analytical technology (PAT), polymers, and imaging (16).

We carried out a number of innovative studies on applications of NIR spectroscopy to physical chemistry (16-19). Ikehata et al. investigated thermal phase behavior of triethylamine (TEA)–water mixtures with various concentrations by use of NIR spectroscopy (19). Based on this study they suggested that the microscopic association of non-polar moieties acts as a trigger for the LCST (Lower Critical Solution Temperature)-type phase separation. We (Šašić and Segtnun et al.) applied NIR spectroscopy to study the structure of water (20). To analyze the temperature-dependent variations of water spectra we used principle component analysis (PCA), self-modeling curve resolution (SMCR), and 2D-COS. The PCA revealed that more than 99% variability in the spectra is due to two bands at 6707 and 7082 cm^{-1} arising from the weakly and strongly hydrogen-bonded water species. All the spectral analysis methods mentioned above supported the two-state model of water, although the model is gradually degraded as temperature decreases (20). Based on the IR and NIR spectra measurements and their quantum chemical calculations Futami et al. showed that it is possible to differentiate hydrogen bonding effects from solvent effects by comparing a change in absorption intensity of a NH or OH fundamental with that of the corresponding overtone (21). Figure 3 compares the effect of hydrogen bonding between fundamentals and first overtones.

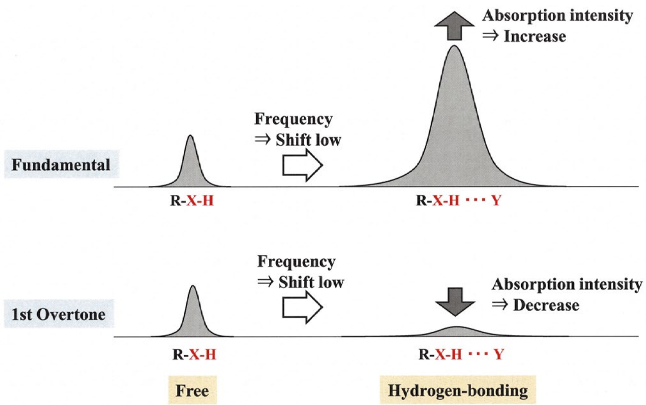


Figure 3. Comparison of the effect of hydrogen bonding between fundamentals and first overtones.

Moreover, we developed a new NIR spectroscopic method based on SPR called light absorption responsive SPR-NIR spectroscopy to aim at dramatic sensitivity improvement (22). The SPR-NIR system which we developed employed an attachment of Kretschman configuration equipped with a mechanism for fine angular adjustment of incident light. This technique may be useful not only for a highly sensitive detection but also for a detection of small amount of samples. This study may lead to develop enhanced NIR spectroscopy.

We have also played important roles in developing NIR imaging. We have been concerned with the development of two kinds of NIR imaging instruments (23); one is a highly sensitive portable NIR imaging device named D-NIRs

(Yokogawa Electric Co.), which is a compact (151×93×120 mm3) instrument. Another is an NIR instrument with high-speed, wide-area monitoring having a highly sensitive NIR camera, named Compovision (Sumitomo Electric Industries), Moreover, Ishimaru et al. developed an imaging-type two-dimensional Fourier spectroscopy (ITFS) system.

Ishikawa et al. carried out innovative NIR imaging studies of polymers and pharmaceutical tablets by D-NIRs and Compovision (23). For example, they studied an aqueous dispersion into a tablet using the latter instrument (23). Ishigaki et al. investigated the growth process of fertilized eggs from Japanese medaka (a freshwater fish) non-invasively using NIR spectroscopy and NIR imaging (24a). The NIR imaging system used in this research, which holds a hyperspectral camera equipped with an InGaAs photodiode array, enables one to acquire microscopic imaging data only in a few seconds.

Ishigaki et al. also carried out nonstaing blood flow imaging using optical interface due to Doppler shift (ITFS) (24). Obtaining nonstaining blood flow images using heterodyne optical interference and images of molecular distribution using molecular vibrational information simultaneously demonstrates an exciting advance in NIR imaging.

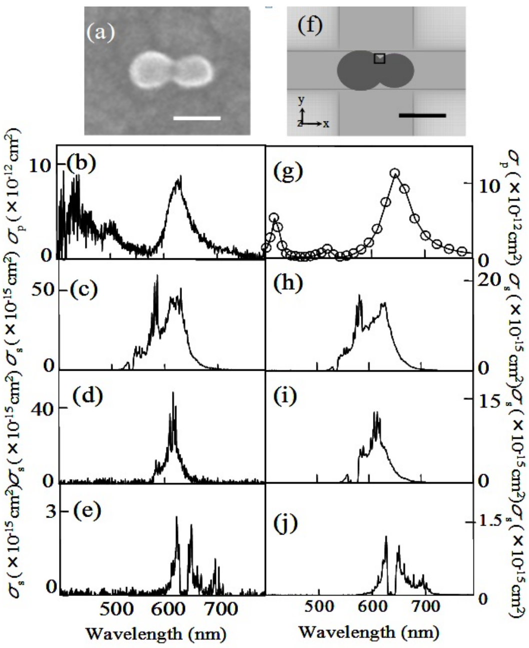
4. Investigations of mechanism of surface-enhanced Raman scattering (SERS) and tip-enhanced Raman scattering (TERS) and their applications to physical chemistry, analytical chemistry, and nanomaterials chemistry

(a) Investigations of mechanism of SERS: We (Itoh et al.) focused on electromagnetic enhancement mechanism as general mechanism of SERS and carried out its research (25,26). Before we started our research on this topic SERS mechanism had been investigated using an ensemble nanoparticles system. In such system, the cause of the enhancement (plasmon resonance) is averaged, and thus, it was difficult to explore directly the relation between the cause and the result (SERS). We solved this problem by use of a single nanoparticle system. We designed a novel experimental system that enabled us to carry out electron microscope measurement, plasmon resonance measurement, and SERS measurement for the same single nano particle dimer. Using the experimental results obtained by this experimental setup as the calculation conditions for electromagnetic analysis, we reproduced SERS cross section. By comparing the experimental SERS spectra with the calculated SERS spectra, we succeeded in quantitative verification of electromagnetic enhancement effect. By this study we contributed markedly to the advances in SERS (Figure 4) (25,26).

Figure 4. Comparison of the experimental (a-e) and calculated results (f-j). (a),(f) dimers of silver nano particles. (b),(g) Experimental and calculated plasmon resonance spectra. (c-e), (h-j) Experimental and calculated SERS spectra excited with 532, 561, and 633 nm. (ref.26)

We are a rather rare group who has explored the electromagnetic mechanisms (25,26), chemical mechanism (27), and that of semiconductor-enhanced Raman scattering (28). Studies of semiconductor-enhanced Raman scattering were started in 1982 by Prof. Haruka Yamada whom I succeeded (29). Ji et al. has worked on the mechanism of semiconductor enhanced Raman scattering (28). Recent our study on semiconductor SERS revealed that Mie resonances scattered near-field effect provided a coherence framework for modeling the electromagnetic mechanism of SERS on semiconductors (28).

(b) Applications of SERS and TERS to physical chemistry, analytical chemistry, and nanomaterials chemistry: We started applications of SERS in 1990s. One of our important SERS papers in 1990s was a report on “indirect SERS” (30). We explored enzyme immunoassay utilizing indirect SERS of the enzyme reaction product. Another interesting study was a SERS study of quantitative analysis of double-stranded DNA amplified by PCR (31). At that time, SERS of DNA was rather rare. Recently, we performed two important SRES immunoassay studies; one is “Immunoassay using probe-labelling immunogold nanoparticles with silver staining enhancement via surface-enhanced Raman scattering” (32a) and another is “label-free indirect immunoassay using an avidin-induced SERS substrate” (32b).



For the last 10 years so we have focused on label-free protein detections by SERS (33). One of our interesting studies is “Coupling reaction-based ultrasensitive detection of phenolic estrogens using surface-enhanced resonance Raman scattering” (34a). Another recent noticeable study is “nickel nanowires combined with SERS: application in label-free detection of cytochrome c-mediated apoptosis” (34b). Rapid label-free Cyt c quantification can be achieved by SERS with a limit of detection of 1 nM. Moreover, a recent investigation on the application of SERS to enantioselective discrimination of alcohols has been highly noted (35). A 3D SERS imaging study of polymers has also received keen interest (36).

We carried out unique studies of TERS. For example, we designed a chemically modified TERS tip for liquid TERS (37). Using this tip, nanoscale quantitative measurement of pH profile at a solid-liquid interface has been made. TERS was also employed in our group to characterize step, ridge, and crack submicro/nanostructures of epitaxial graphene on 4H-SiC $(000\bar{1})$ (38). We also developed a high vacuum low temperature TERS system with Unisoku Co. Ltd. to explore nanomaterials without effects of atmosphere and thermal fluctuation.

5. FIR/Terahertz and low-frequency Raman spectroscopies and quantum chemistry studies of polymers

One can say three spectroscopies, FIR, THz, and low-frequency Raman spectroscopies form low-frequency vibrational spectroscopy. In the low-frequency region bands due to stretching modes including heavy atoms, skeletal vibrations, twisting vibrations, lattice vibrations, and intermolecular vibrations appear. Band assignments in the low-frequency region are, in general, not easy. Low-frequency vibrational spectra of molecules often reflect their intramolecular and intermolecular interactions, and therefore, they have recently been utilized to investigate higher order structures of polymers and proteins, their intermolecular interactions, hydrogen bondings, hydrations and dynamics.

Our strategy for polymer research by low-frequency vibrational spectroscopy is to compare FIR/THz, and low-frequency Raman spectra and carry out quantum mechanical calculation combined with the Cartesian coordinate tensor transfer (CCT) method for band assignments (6,7). THz, FIR and Raman spectra including polarized spectra of two kinds of biodegradable polymers, poly-(R)-3-hydroxybutyrate (PHB) and poly (glycolic acid) (PGA) having weak hydrogen bondings and nylon 6 with strong hydrogen bondings were investigated (6,7). The intermolecular interactions among the polymer chains were considered explicitly in our DFT calculations aided by fragmentation methodology (CCT method).

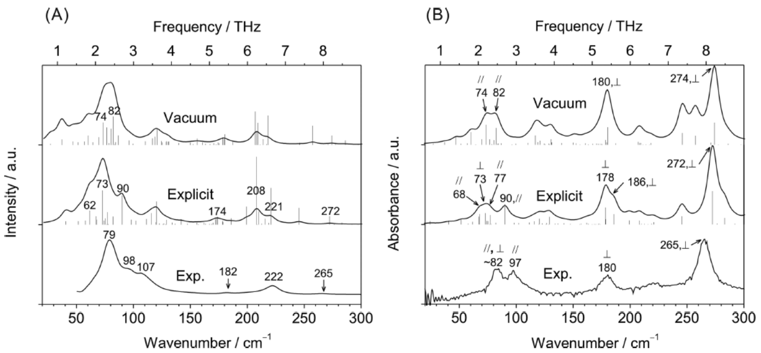


Figure 5. Bottom in (A) and (B); Raman and FIR absorption spectra of crystalline PHB, respectively. Top and middle in (A) and (B); their calculated spectra (ref.6).

Figure 5 bottom in (A) and (B) depict Raman and FIR absorption spectra of crystalline PHB, respectively, and top and middle in (A) and (B) show their calculated spectra (6). The calculations were carried out under vacuum (top) and with the explicit correction for the intermolecular interactions (middle). Of note is that the main features of both Raman and FIR spectra of PHB were reproduced by both calculations irrespective of the intermolecular interactions were considered or not. However, the explicit correction yields much better agreements between the experiments and the calculations in terms of the spectral shapes, relative intensities, and relative frequencies. Moreover, the explicit calculations and the experiments in both of the Raman and FIR spectra led Yamamoto et al. to propose the band assignments; they assigned the Raman bands at 79 and 98 cm^{-1} to those at 73 (out-of-plane C= O + CH_3) and 90 (CH_2 + CH_3) cm^{-1} in the explicit calculation (6).

There were a lot of dispute on assignments of the low-frequency modes of nylon. Our study has provided them with defined assignments. We found a similarity in 125 and 70 cm^{-1} regions between crystalline polyesters and nylon 6 that both polymers show specific out-of-plane vibrational peaks around 100 cm^{-1}, which are sensitive to the lattice length among polymer chains (6,7).

6. Medical Applications of Raman and NIR Spectroscopy and Applications of Raman Spectroscopy to Basic Biology

I am one of the pioneers in medical Raman spectroscopy (39). I together with started my nondestructive Raman study of the mechanism of cataract formation in 1981 (40). This may be the first application of Raman spectroscopy to an investigation of disease mechanism. This study demonstrated the potential of Raman spectroscopy in medical diagnosis. I also made significant contributions to medical applications of NIR spectroscopy and applications of Raman spectroscopy to basic biology.

(a) Medical applications of Raman spectroscopy: In 1980s I together with Mizuno et al. proceeded the studies of cataracts and those of lens aging in parallel using mouse and rat lenses (39,40). We found nondestructively the significant difference in the mechanism of cataractogenesis and lens aging at the molecular level. In the case of mouse lens aging the water content in a lens decreases gradually while that in a cataract lens increases. Moreover, during cataract formation some of buried tyrosine residues of lens proteins become exposed (40). Figure 6 (A) and (B) compare age-dependent Raman spectral variations in the 950-700 cm^{-1} region (tyrosine doublet region) of a normal rat lens and a cataract rat lens, respectively. Note that the intensity ratio of the tyrosine doublet (855/832 cm^{-1}) changes for the cataract lens. I wrote a very important review paper on Raman applications to medical studies in 1988 (39).

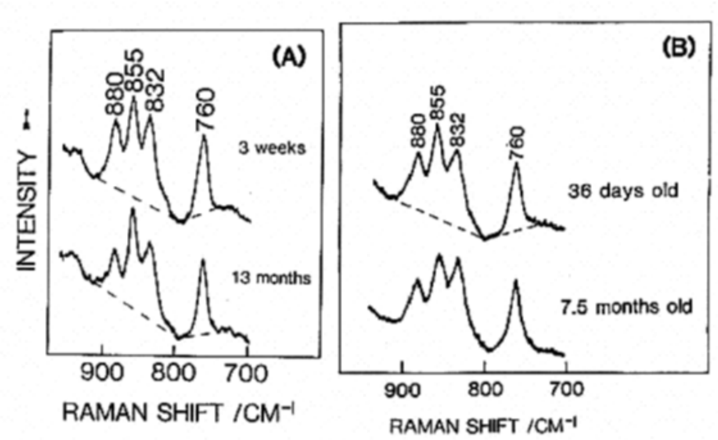


Figure 6. Comparison of age-dependent Raman spectral variations in the 950-700 cm^{-1} region (tyrosine doublet region) of (A) a normal rat lens and (B) a cataract rat lens (ref.40).

In 1992 we used 1064-nm excitation FT-Raman spectroscopy to measure Raman spectra of brain tissues. Subsequently, we reported Raman spectra of brain cancer tissues and found various spectral variations for the tissues due to cancer (41). Glioma grade III showed a strong band at 856 cm^{-1} due to polysaccharides. One of the neurinoma contained a carotenoid which exhibited two resonance Raman peaks at 1157 and 1524 cm^{-1}. The appearance of a strong Raman peak at 960 cm^{-1} suggested that the central neurocytoma of choroid plexus was calcified. These are examples of initiatives of cancer tissue Raman studies.

Recently, we (Ishigaki et al.) investigated esophageal cancer in an early stage by use of Raman spectroscopy. Of note in this study was that a kind of neural network was used to analyse Raman spectra for diagnosis of the cancer tissues (42). We also carried out a Raman imaging study of cancer tissues (43). We compared several chemometrics methods for extracting the contribution from paraffin of cancer tissue samples in Raman imaging measurement.

(b) Applications of Raman spectroscopy to basic biology: Recently, many Raman spectroscopists are concerned with medical or biomedical applications of Raman spectroscopy, and also its applications to biochemistry are active. However, relatively few research groups have been involved in its applications to fundamental and basic biology. Therefore, we pay attention to their applications. For example, using Raman spectroscopy Ishigaki et al. investigated mouse embryo development and its qualitative evaluation at the molecular level in a nondestructive manner (44). A Raman spectra measured in situ during embryonic development revealed that proteins with a β-sheet structure are derived from maternal oocytes, while α-helical proteins are additionally generated by switching on a gene after fertilization. We also explored potential biomarkers indicating developmental competence of matured oocytes by Raman spectroscopy (44). It was found that phosphoric acid and phosphorylation levels are potential biomarkers. These studies have opened a new area of applications of Raman spectroscopy to fundamental biology.

(c) Medical applications of NIR spectroscopy: I together with Matsunaga began our biomedical applications of NIR spectroscopy at the end of 1980's. We succeeded in measuring NIR spectra of blood hemoglobin in a human hand in a nondestructive manner (45). We could monitor noninvasively a change of oxyhemoglobin in the blood. Later, this study was extended to our NIR noninvasive observation of blood sugar in an arm. For this purpose, I aided a Panasonic group to develop a specialized NIR spectrometer with high sensitivity and low noise. Moreover, we performed NIR spectroscopic determination of human serum albumin, γ-globulin, and glucose in a control serum solution with newly developed chemometrics algorithms (searching combination moving window partial least squares) (46).

As stated, above we have challenged to unexplored undeveloped fields of molecular spectroscopy with original idea and methods and developed markedly them. We have contributed significantly to modern molecular spectroscopy through the establishment of basic principles, the development of instruments, the proposals for the novel spectral analysis methods and the applications of quantum chemistry to molecular spectroscopy. Our achievements have been internationally recognized and have had a strong impact on a wide research area not only chemistry but also biology and medicine.

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Surface-enhanced Raman scattering (SERS) of chemisorbed species on various kinds of metals and semiconductors, *Surf. Sci.,* 134, 71 (1983).

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(43) P. Meksiarun, M. Ishigaki, V.A.C. Huck-Pezzei, C.W. Huck, K. Wongravee, H. Sato, Y. Ozaki, Comparison of multivariate analysis methods for extracting the paraffin component from the paraffin-embedded cancer tissue spectra for Raman imaging. *Sci. Rep*. 7, 44890 (2017).

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(45) Y. Ozaki, T. Matsunaga, T. Miura: Nondestructive and Noninvasive Monitoring of Deoxyhemoglobin in the Vein by Use of a Near-Infrared Reflectance Spectrometer with a Fiber-Optic Probe, *Appl. Spectrosc.,* 46**,** 180 (1992).

(46) Y. P. Du, Y. Z. Liang, J. H. Jiangc, R. J. Berry, Y. Ozaki; Spectral regions selection to improve prediction ability of PLS models by changeable size moving window partial least squares and searching combination moving window partial least squares *Anal. Chim. Acta*, 501, 183 (2004).

3. Academic Activities

Editorial members

1. Encyclopedia of Analytical Chemistry, Associate editor
2. Vibrational Spectroscopy, editorial member
3. Journal of Raman Spectroscopy, editorial member
4. Journal of Molecular Structure, editorial member
5. Journal of Near Infrared Spectroscopy, editorial member
6. Chinese Journal of Light Scattering, editorial member
7. Spectroscopy and Spectral Analysis, editorial member

Organizing committee members of international conferences

1. International Conference on Advanced Vibrational Spectroscopy (one-time chair and organizing committee member）
2. UV Photonics of SPIE (organizing committee member)
3. International Conference on Raman Spectroscopy (one-time organizing committee member; 2012-2018)
4. Pacifichem symposium chair, 2010, 2015, 2020
5. FACSS (Federation of Analytical Chemistry and Spectroscopy Societies (one-time representative of Japan; 2014-2018）
6. The Asian NIR Consortium (ANC)（one-time president; 2009-2013）
7. SciX session chair (2014-)

Awards and Distinctions

International Awards

(1) Tomas Hirschfeld Award (1998). International Consortium for Near Infrared Spectroscopy

(2) EAS Award in Achievement in Near-Infrared Spectroscopy (2001), Eastern Analytical Symposium. USA

(3) The Büchi NIR Award (2002), Büchi. Switzerland

(4) Gerald S. Birth Award (2006), The Council of Near Infrared Spectroscopy

(5) Dasari Lecture Award (2011), George R. Harrison Spectroscopy Laboratory, MIT

(6) Bomem-Michelson Award (2014). The Coblentz Society, USA

(7) China Friendship Award on Molecular Spectroscopy (2016). The Chinese Optical Society

(8) Lu Wanzhen Memorial China Instrument and Control Society Award for International Exchange (2018).

(9) Memorial Award for the 90th Anniversary of the Discovery of Raman Effect (2018). Bangalore, India

(10) Pittsburg Spectroscopy Award (2019), Pittsburg Spectroscopy Society. USA

Domestic Awards

(1) The Young Scientist Award (1987). The Spectroscopical Society of Japan.

(2) The Spectroscopical Society of Japan Award (2002).

(3) Hyogo Prefecture Science Award (2003).

(4) Science and Technology Award of Ministry of Education, Culture, Sports, Science and Technology (2005).

(5) Japan Analytical Chemistry Society Award (2008).

(6) Japan Analytical Chemistry Society Advanced Analytical Technique Award (2011).

(9) The Chemical Society of Japan Award (2017).

(10) The Medal with Purple Ribbon from Japanese Emperor (2018).

Distinctions and Honors

(1) Honorary Professor; Jilin University, Changchun, China (2007-).

(2) Honorary Professor; Changchun Institute of Applied Chemistry, Changchun, China (2007-).

(3) Guest Professor; Peking University, Peking, China (2009-).

(4) Gold Medal Award, University of Wroclaw, Poland (2009).

(5) Changbai Mountain Friendship Award, China (2010).

(6) Dr. Ramachandra Rao Dassari Distinguished Lecture (Indian Institute of Technology, Kanpur) (2012).

(7) Jagiellonian University 650th Anniversary PLUS RATIO VIS Silver Medal (2014).

(8) Doctor Honoris Causa, Jagiellonian University, Poland (2016).

(9) Guest Professor, University of Innsbruck. Austria (2018).

(10) Doctor Honoris Causa, University of Wroclaw, Poland (2019).

Fellows

(1) Fellow, Society for Applied Spectroscopy (2010).

(2) Fellow, The Royal Society of Chemistry (2015).

(3) Fellow, The Chemical Society of Japan (2016).

(4) Fellow, International Council of Near-infrared Spectroscopy (2017).

4. List of Publications (2013-Feb. 2020）

(1) T. Suzuki, X. Yan, Y. Kitahama, H. Sato, T. Itoh, T. Miura, Y. Ozaki: Tip-Enhanced Raman Spectroscopy Study of Local Interactions at the Interface of Styrene−Butadiene Rubber/Multiwalled Carbon Nanotube Nanocomposites, The Journal of Physical Chemistry, C, 117, 1436-1440 (2013)

(2) K. Wongravee, T. Parnklang, P. Pienpinijtham, C. Lertvachirapaiboon,Y. Ozaki, C. Thammacharoena and S.

Ekgasita: Chemometric analysis of spectroscopic data on shape evolution of silver nanoparticles induced by hydrogen peroxide, Phys. Chem. Chem. Phys., 15, 4183-4189 (2013).

(3) Hideyuki Shinzawa, Kimie Awa, Isao Noda, Yukihiro Ozaki: Pressure-induced variation of cellulose tablet studied by two-dimensional (2D) near-infrared (NIR) correlation spectroscopy in conjunction with projection pretreatment. Vibrational Spectroscopy, 65, 28-35 (2013).

(4) Shigeki Yamamoto, Yusuke Morisawa, Harumi Sato, Hiromichi Hoshina, and Yukihiro Ozaki: Quantum Mechanical Interpretation of Intermolecular Vibrational Modes of Crystalline Poly-(R)-3-Hydroxybutyrate Observed in Low- Frequency Raman and Terahertz Spectra, J. Phys. Chem. B, 117, 2180-2187 (2013).

(5) Chihiro Hashimoto, Akiyoshi Nagamoto, Takashi Maruyama, Naomi Kariyama, Yuma Irisa, Akifumi Ikehata, and Yukihiro Ozaki: Hydration States of Poly(N-isopropylacrylamide) and Poly(*N,N*-diethylacrylamide) and Their Monomer Units in Aqueous Solutions with Lower Critical Solution Temperatures Studied by Infrared Spectroscopy, Macromolecules, 46(3), 1041-1053 (2013).

(6) Hideyuki Shinzawa, Kimie Awa, Isao Noda, Yukihiro Ozaki , Multiple-Perturbation Two-Dimensional Near-Infrared Correlation Study of Time-Dependent Water Absorption Behavior of Cellulose Affected by Pressure, Applied Spectroscopy, 67,163-170 (2013).

(7) Kodai Murayama, Takuma Genkawa, Daitaro Ishikawa, Makoto Komiyama, and Yukihiro Ozaki: A polychromator-type near-infrared spectrometer with a high-sensitivity and high-resolution photodiode array detector for pharmaceutical process monitoring on the millisecond time scale: Review of Scientific Instruments, 84, 023104-1-8 (2013).

(8) Miriam Unger, Harumi Sato, Yukihiro Ozaki, Dieter Fischer, Heinz W. Siesler: Temperature-Dependent Fourier Transform Infrared Spectroscopy and Raman Mapping Spectroscopy of Phase-Separation in a Poly(3-hydroxybutyrate)–Poly(L-Lactic Acid) Blend: Applied Spectroscopy, 67, 2, 141-147(2013).

(9) Yasutaka Kitahama, Masato Kashihara, Tamitake Itoh, and Yukihiro Ozaki: Surface-Enhanced Phosphorescence Measurement by an Optically Trapped Colloidal Ag Nanoaggregate on Anionic Thiacarbocyanine H-Aggregate: J. Phys. Chem. C, 117, 2460−2466 (2013).

(10) Yuta Miyamae, Marie Kawabata, Yumika Yamakawa and Yukihiro Ozaki: Review article: Non-invasive assessment for photoaging and physiological aging of human skin and non-invasive estimation of the thickness of mice skin by near infrared diffuse-reflectance spectroscopy: NIR news, 24, 1, 11-14, (2013).

(11) Shigeki Yamamoto, Yusuke Morisawa, Harumi Sato, Hiromichi Hoshina and Yukihiro Ozaki: Quantum Mechanical Interpretation of Intermolecular Vibrational Modes of Crystalline Poly-(R)-3-Hydroxybutyrate Observed in Low- Frequency Raman and Terahertz Spectra: J. Phys. Chem., B, 117, 2180-2187 (2013).

(12) Dariusz Sobolewski, Edyta Proniewicz, Dominika Skołuba, Adam Prahl, Yukihiro Ozaki, Younkyoo Kim and Leonard M. Proniewicz: Characterization of adsorption mode of new B2 bradykinin receptor antagonists onto colloidal Ag substrate: Journal of Rman Spectroscopy,44, 212-218(2013).

(13) Edyta Proniewicz, Prompong Pienpinijtham, Yukihiro Ozaki, Younkyoo Kim and Leonard M. Proniewicza: Influence of backbone length and synthetic mutations on orientation of neurotensin fragments adsorbed onto a colloidal silver surface: SERS studies: Journal of Rman Spectroscopy, 44, 55-62(2013).

(14) Akifumi Uda, Shigeaki Morita, Yukihiro Ozaki: Thermal degradation of a poly(vinyl alcohol) film studied by

multivariate curve resolution analysis: Polymer, 54, 2130-2137(2013).

(15) Xinlei Yan, Tamitake Itoh, Shouyu Dai, Yukihiro Ozaki, Yan Fang: Cu,Mu doping effect to optical behavior and electronic structure of ZnO ceramic: Journal of Physics and Chemistry of Solids, 74, 1127-1130 (2013).

(16) Edyta Proniewicz, Natalia Piergies, Yukihiro Ozaki, Younkyoo Kim, Leonard M. Proniewicz: Investigation of adsorption mode of a novel group of N-benzylamino (boronphenyl) methylphosphonic acids using SERS: Molecular and Biomolecular Spectroscopy, 103, 167-172(2013).

(17) Xiuxiang Gao, Yufeng Liu, Huizhen Li, Jiang Bian, Ying Zhao, Ye Cao, Yuezhi Mao, Xin Li, Yizhuang Xu, Yukihiro Ozaki, Jinguang Wu: A cooperative hydrogen bonding system with a CAH…O hydrogen bond in ofloxacin: Journal of Molecular Structure,1040, 122–128 (2013).

(18) Takeyoshi Goto, Akifumi Ikehata, Yusuke Morisawa, and Yukihiro Ozaki: Electronic Transitions of Protonated and Deprotonated Amino Acids in Aqueous Solution in the Region 145−300 nm Studied by Attenuated Total Reflection Far-Ultraviolet Spectroscopy: J. Phys.Chem. , 117, 2517−2528 (2013).

(19) Hiromichi Hoshina, Shinya Ishii, Shigeki Yamamoto, Yusuke Morisawa, Harumi Sato, Tetsuji Uchiyama, Yukihiro Ozaki, and Chiko Otani: Terahertz Spectroscopy in Polymer Research: Assignment of Intermolecular Vibrational Modes and Structural Characterization of Poly(3-Hydroxybutyrate), IEEE Transactions on Terahertz Science and Technology, 3, 248-258 (2013)

(20) Takeyoshi Goto, Yusuke Morisawa, Noboru Higashi, Akifumi Ikehata, and Yukihiro Ozaki: Pulse Laser Photolysis of Aqueous Ozone in the Microsecond Range Studied by Time-Resolved Far-Ultraviolet Absorption Spectroscopy: Analytical Chemistry, 85, 4500-4506 (2013).

(21) Yeonju Park, Chihiro Hashimoto, Takeji Hashimoto, Yoshitsugu Hirokawa, Young Mee Jung and Yukihiro Ozaki: Reaction-Induced Self-Assembly of Gel Structure: A New Insight into Chemical Gelation Process of N-Isopropylacrylamide as Studied by Two-Dimensional Infrared Correlation Spectroscopy: Macromolecules, 46, 3587-3602 (2013).

(22) Tamitake Itoh, Yuko S. Yamamoto, Hiroharu Tamaru, Vasudevanpillai Biju, Norio Murase, and Yukihiro Ozaki: Exciation laser energy dependence of surface-enhanced fluorescence showing Plasmon-induced ultrafast electronic dynamics in dye molecules: Physical Review B, B 87, 235408 (2013)

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(24) Tamitake Itoh, Yuko S. Yamamoto, Hiroharu Tamaru, Vasudevanpillai Biju, Norio Murase and Yukihiro Ozaki: Excitation Laser energy dependence of surface-enhanced fluorescence showing Plasmon-induced ultrafast electronic dynamics in dye molecules: Physical Review., B87, 235408 (2013)

(25) Edyta Proniewicz, Yukihiro Ozaki, Younkyoo Kim and Leonard M. Proniewicz: Adsorption mode of neurotensin family peptides onto a colloidal silver surface: SERS studies: J Raman Spectrosc., 44, 355-361 (2013)

(26) Takuma Genkawa, Masahiro Watari, Takashi Nishii, Masao Suzuki and Yukihiro Ozaki: Two-Dimensional Heterospectral Correlation Analysis of Water and Liquid Oleic Acid Using an Online Near-Infrared/Mid-Infrared Dual-Region Spectrometer: Applied Spectroscopy, 67, 724-730 (2013)

(27) Wei Ji, Lei Chen, XiangxinXue, Zhinan Guo, Zhi Yu, Bing Zhao and Yukihiro Ozaki: Design of an anti-

aggregated SERS sensing platform for metal ion detection based on bovine serum albumin-mediated metal nanoparticles: The Royal Society of Chemistry, 49, 7334-7336 (2013).

(28) Edyta Proniewicz, Natalia Piergies, Yukihiro Ozaki, Younkyoo Kim and Leonard M. Proniewicz: Influence of Substituent Type and Position on the Adsorption Mechanism of Phenylboronic Acids: Infrared, Raman, and Surface Enhanced Raman Spectroscopy Studies: J. Phys. Chem., 117, 5693-5705 (2013).

(29) Daitaro Ishikawa, Kodai Murayama, Kimie Awa, Takuma Genkawa, Makoto Komiyama, Sergei G. Kazarian and Yukihiro Ozaki: Application of a newly developed portable NIR imaging device to monitor dissolution process of tablets: Springer, vol.405, No.29, p.9401-9409, (2013).

(30) Xinlei Yan, Toshiaki Suzuki, Yasutaka Kitahama, Harumi Sato, Tamitake Itoh and Yukihiro Ozaki: A study on the interaction of single-walled carbon nanotubes (SWCNTs) and polystyrene (PS) at the interface in SWCNT-PS nanocomposites using tip-enhanced Raman spectroscopy: Phys. Phys. Chem. Chem. Phys., 15, 20618-20624(2013).

(31) Daitaro Ishikawa, Shinya Fukuda, Shigenori Arimitsu, Kazuhiko Ohba, Yukihiro Ozaki and Etsuji Ishiguro: A case study on evaluation of water contents change in leaves(Sudajii) by using chlorophyll absorption band in the 400-1100 nm region: J. Agric.Meteorol., 69, 201-207 (2013).

(32) Daitaro Ishikawa, Takashi Nishii, Fumiaki Mizano, Harumi Sato, Sergei G. Kazarian, Yukihiro Ozaki: Potential of a Newly Developed High- Speed Near- Infrared (NIR) Camera (Compovision) in Polymer Industrial Analyses: Monitoring Crystallinity and Crystal Ebolution of Polylactic Acid (PLA) and Concentration of PLA in PLA/Poly-(R)-3-Hydroxybutyrate(PHB) Blends: Applied Spectroscopy, 67, 1441-1446 (2013)

(33) Yuko S. Yamamoto, Katsuyuki Hasegawa, Yuuki Hasegawa, Naoshi Takahashi, Yasutaka Kitahama, Satoshi Fukuoka, Norio Murase, Yoshinobu Baba, Yukihiro Ozaki and Tamitake Itoh: Direct conversion of silver complexes to nanoscale hexagonal columns on a copper alloy for plasmonic applications: Physical Chemistry Chemical Physics, 15, 14611-14615 (2013).

(34) Daitaro Ishikawa, Takashi Nishii, Fumiaki Mizuno, Sergei G.Kazarian and Yukihiro Ozaki: Development of a high- speed monitoring near infrared hyperspectral camera (compovision) for wide area imaging and its applications: NIR news (2013)

(35) Yusuke Morisawa, Manaka Yasunaga, Ryoichi Fukuda, Masahiro Ehara and Yukihiro Ozaki: Electronic transitions in liquid amides studied by using attenuated total reflection far-ultraviolet spectroscopy and quantum chemical calculations: The Journal of Chemical Physics, 139, UNSP154301 (2013).

(36) T. Suzuki, X. Yan, Y. Kitahama, H. Sato, T. Itoh, T. Miura, Y. Ozaki: Tip-Enhanced Raman Spectroscopy Study of Local Interactions at the Interface of Styrene - Butadiene Rubber/Multiwalled Carbon Nanotube Nanocomposites, The Journal of Physical Chemistry, The Journal of Physical Chemistry C, 117, 1436-1440 (2013).

(37) Yujing Chen,ab Yukihiro Ozaki and Miroslaw A. Czarnecki: Molecular structure and hydrogen bonding in pure liquid ethylene glycol and ethylene glycol water mixtures studied using NIR spectroscopy: Phys. Chem. Chem. Phys., 15, 18694-18701 (2013)

(38) Caihong Zhang, Xinlei Yan, Liyuan Wang, Yasutaka Kitahama, Yukihiro Ozaki et al: The effect of temperature on the resonance of the interband transition energy in single-walled carbon nanotubes with excitation laser energy

by Raman spectroscopy, Appl. Phys. Lett. 103, 231902 (2013)
(39) D.Ishikawa, G. Hoogenboom, Y. Ozaki and E. Ishiguro: A Study on the Spectral Change in a Chlorophyll Absorption Band Monitored During the Growth of Japanese Tea Leaves, J. Agric Meteorol , 69(4), 255 (2013)
(40) Yuko S. Yamamoto, Mitsuru Ishikawa, Yukihiro Ozaki and Tamitake Itoh: Fundamental studies on enhancement and blinking mechanism of surface-enhanced Raman scattering (SERS) and basic applications of SERS biological sensing: Front. Phys, 9, 31-46 (2014).
(41) Atsushi Kuriyama and Yukihiro Ozaki: Assessment of Active Pharmaceutical Ingredient Particle Size in Tablets by Raman Chemical Imaging Validated using Polystyrene Microsphere Size Standards: AAPS PharmSciTech, 15, 375-387 (2014)
(42) Ichiro Tanabe and Yukihiro Ozaki: Consistent changes in electronic states and photocatalytic activities of metal (Au, Pd, Pt)-modified TiO2 studied by far-ultraviolet spectroscopy: ChemComm., 50, 2114-2119 (2014)
(43) Kimie Awa, Hideyuki Shinzawa and Yukihiro Ozaki: An Effect of Cellulose Crystallinityon the MoistureAbsorbability of a Pharmaceutical Tablet Studied by Near-Infrared Spectroscopy, Applied Spectroscopy, 68(6), 625 (2014)
(44) Yujing Chen, Yusuke Morisawa, Yoshisuke Futami, Mirostaw A. Czarnecki, Hai-Shi Wang and Yukihiro Ozaki: Combined IR/NIR and Density Functional Theory Calculations Analysis of the Solvent Effects on Frequencies and Intensities of the Fundamental and Overtones of the C O Stretching Vibrations of Acetone and 2-Hexanone, J. Phys. Chem.,118, 2576 (2014)
(45) Ichiro Tanabe, Takayuki Ryoki and Yukihiro Ozaki: Significant Enhancement of Photocatalytic Activity of Rutile TiO2 Compared with Anatase TiO2 upon Pt Nanoparticle Deposition Studied by Far-Ultraviolet Spectroscopy, Phys. Chem. Chem. Phys., 16, 7749(2014)
(46) Yoshisuke Futami, Yasushi Ozaki, Yoshiaki Hamada, Yukihiro Ozaki: Frequencies and absorption intensities of the fundamental and the first overtone of NH stretching vibrations of pyrrole– acetylene and pyrrole– ethylene complexes studied by density – functional – theory calculation, Vibrational Spectroscopy, 72, 124-127 (2014)
(47) Hideki Shinzawa, MasakazuNishida, AkiraTsuge, DaitaroIshikawa, YukihiroOzaki, Shigeaki Morita, Wataru Kanematsu: Thermal Behavior of Poly(lactic acid)-Nanocomposite Studied by Near-InfraredImaging Based on Roundtrip Temperature Scan, Appl. Spectrosc. 68(3), 371 (2014)
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2020/07/17

List of Publications

Yukihiro Ozaki

July. 12, 2020

PART A: Original Papers and Review Papers

PART B: Book Chapters

PART C: Books

Total number of original papers and review papers, **1036**, H-Index, **74**, Total number of citations, **26,000**

PART A, Original Papers and Review Papers

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