高次複合光応答分子システムの開拓と学理の構築

領域番号：2606

略称：高次複合光応答

平成２６年度～平成３０年度

科学研究費助成事業（科学研究費補助金）

（新学術領域研究（研究領域提案型））

研究成果報告書

令和２年６月

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はじめに

平成２６年度にスタートした新学術領域研究「高次複合光応答」は、５年間の研究期間を終え、平成３１年３月に終了いたしました。

電子励起状態分子はエネルギーや物質変換、光機能発現等において重要な役割を果たしています。しかし、光機能発現やエネルギー・物質変換に応用が想定される比較的大きな分子では、① 高位電子状態が生成しても迅速に最低励起状態に緩和し（Kasha 則）（光量子の質の損失)、② 集合体系において多くの励起分子が生成しても分子間の迅速な消滅過程（励起子 annihilation）により最終的にはごく少数の励起分子しか残らない（光量子の量の損失） など、光子の質や量の損失に関わる大きな制限が普遍的に存在します。更に、③ 通常の光吸収で到達できる電子状態は主に１光子許容なものに限られており、分子が本来有する多様な電子励起状態を有効に利用することも困難でした（多様性の損失)。

これらの制限のために、分子・物質系の電子状態選択的な応答、多数の励起分子の反応やその応答が協動する光応答、これらの結果として進行するメゾスコピック・マクロスコピックスケールの分子集合体系の光応答・機能などへの高次展開は困難なものとされ、主な研究対象は“１光子吸収による最低励起状態からの１分子の光応答”に限られていました。

これらの背景を基に、本新学術領域では、多重・多光子励起、電子状態変調、分子・集合体開発 等の方法を用い、従来の“１光子吸収と１分子応答”を超えた光子有効利用を可能とす複合光応答（英語では Photosynergetics と標記）系の開拓・発展により、今後の解決すべき分子系の光利用関連諸課題の共通基盤の構築を目標としました。

この間、物理化学、理論化学、材料化学、有機化学、無機化学、物性物理、デバイス作成など広範な研究者の参画を得て、分子・集合体・巨視的物質系における多数光子・多数励起子（電子励起状態分子）の示す高次複合光応答の開拓、機構解明、発展をめざした研究を行ってきました。これらの結果から、次代の電子励起状態分子の利用に関わる光化学・光物質化学の発展に関わる基礎的な知見、研究の方法論を含めた共通基盤を多数得ることができたと考えています。

この５年間、種々のご支援をいただきましたことに、深く感謝申し上げますとともに、これらの成果を基にして、新たな研究が発展することを期待しています。

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（A02 班）

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交付決定額（配分額）

年度	直接経費	間接経費	計
平成 26 年度	187,900,000	56,370,000	244,270,000
平成 27 年度	214,000,000	64,200,000	278,200,000
平成 28 年度	221,300,000	66,390,000	287,690,000
平成 29 年度	204,400,000	61,320,000	265,720,000
平成 30 年度	196,000,000	58,800,000	254,800,000
計	1,023,600,000	307,080,000	1,330,680,000

単位　（円）

計画研究、公募研究（平成 27-30 年度）、総括班、国際活動支援班（平成 27-30 年度）

研究発表

雑誌論文：　総数 965 本（領域内共同研究による重複を除く）、2019 年 3 月 31 日

以下のリストは研究グループ毎にまとめた。そのため領域内共同研究による共著が重複している。

（査読有り）（総数 931 本）

計画班

A01：宮坂　博

1. M. Koga, Y. Yoneda, H. Sotome, H. Miyasaka, "Ionization Dynamics of a Phenylenediamine Derivative in Solutions as Revealed by Femtosecond Simultaneous and Stepwise Two-Photon Excitation", **Phys. Chem. Chem. Phys.**, 21, 2019, 2889-2898.

2. H. Sotome, D. Kitagawa, T. Nakahama, S. Ito, S. Kobatake, M. Irie, H. Miyasaka, "Cyclization Reaction Dynamics of an Inverse Type Diarylethene Derivative as Revealed by Time-Resolved Absorption and Fluorescence Spectroscopies", **Phys. Chem. Chem. Phys.**, 21, 2019, 8623-8632.

3. T. Nakahama, D. Kitagawa, H. Sotome, S. Ito, H. Miyasaka, S. Kobatake, "Crystallization-Induced Emission of 1,2-Bis(3-methyl-5-(4-alkylphenyl)-2-thienyl)perfluorocyclopentenes: A Mechanical and Thermal Recording System", **Dyes and Pigments**, 160, 2019, 450-456.

4. E. Murakami, R. Mizoguchi, Y. Yoshida, A. Kitashoji, N. Nakashima, T. Yatsuhashi, "Multiple strong field ionization of metallocenes: Applicability of ADK rates to the production of multiply charged transition metal (Cr, Fe, Ni, Ru, Os) cations", **J. Photochem. Photobiol. A**, 369, 2019, 16-24.

5. Y. Ishibashi, M. Arinishi, T. Katayama, H. Miyasaka, T. Asahi, "Femtosecond Excited-State Dynamics of Fullerene-C60 Nanoparticles in Water", **Phys. Chem. Chem. Phys.**, 20, 2018, 958-966.

6. T. Nakahama, D. Kitagawa, H. Sotome, S. Ito, H. Miyasaka, S. Kobatake, "Solid-state fluorescence behavior induced by photochemical ring-opening reaction of 1,2-bis(3-methyl-5-phenyl-2-thienyl)perfluorocyclopentene", **Bull. Chem. Soc. Jpn.**, 91, 2018, 153-157.

7. T. Nakahama, D. Kitagawa, H. Sotome, T. Fukaminato, S. Ito, H. Miyasaka, S. Kobatake, "Fluorescence On/Off Switching in Nanoparticles Consisting of Two Types of Diarylethenes", **ACS Omega**, 3, 2018, 2374-2382.

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学会発表

リストには国際学会における主な招待講演（invited lecture）、基調講演（ketynote lecture）、特別講演（plenary lecture）などを示した。

計画班

1. Hiroshi Miyasaka, “Photosynergetic Responses of Molecules Induced by Multiple Excitation and Multiphoton Absorption”, **10th Asian Photochemistry Conference**, 2018年12月16-20日 Taipei, Taiwan,（特別、受賞）.

2. Hiroshi Miyasaka, “Photosynergetic Responses in Molecules: Multiphoton-Gated Photochromic Reactions”, **French-Japanese International Associated Laboratory Nano-synergetics**, 2018年3月29-30日, Lille, France,（基調）

3. Hiroshi Miyasaka, “Photosynergetic Responses: Multiphoton-Gated Photochromic Reactions”, **Photo-Active Molecules and Materials: Synthesis, Photophysics and Modeling**, 2017年11月30日, Cachan, France, （基調）

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5. Hiroshi Miyasaka, Y. Nagasawa, "Vibrational Wavepacket Motions in Charge Transfer Systems studied by femtosecond ultrafast spectroscopies.", **International Symposium on Pure & Applied Chemistry (ISPAC) 2017**, 2017年6月8日-6月10日, Ho Chi Minh, Vietnam（招待）

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8. Hiroshi Miyasaka, “Anomalous Diffusion of Guest Dyes in Amorphous Polymer Solids as Revealed by Single Molecule Tracking”, **The International Chemical Congress of Pacific Basin Societies 2015** , 2015年12月15-20日, Honolulu, USA（招待）.

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11. T. Katayama, S. Ito, M. Endo, A. Wakamiya, Y. Murata, Y. Ogomi, S. Hayase, H. Miyasaka, "Observation of Inhomogeneous Deactivation Process in Lead Iodide Perovskite-based Solar Cell by Means of Femtoscond Transient Absorption Miroscopy", **8th Asian Photochemistry Conference 2014 (2014APC)**, 2014年11月9日-11月13日, Kerala, India（招待）.

12. Tomoyuki Yatsuhashi, "Organic Nanoparticles Formation by Pulsed-Laser irradiation to liquid/liquid Interfaces", **9th International Conf. on Photoexcited Processes & Applications**, 2014年9月29日-10月3日, Matsue, Japan（招待）

13. Kohei Imura, "Visualization of plasmons in single metal nanoplates using advanced near-field imaging methods", **The 11th Asia-Pacific Conference on Near-Field Optic**, 2017年7月10日-7月13日, Tainan, Taiwan（招待）.

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102. Takashi Hirose, "Design of Highly Photoresponsive 2-D Molecular Orderings Based on Cooperative Assembly at the Liquid/Graphite Interface", **Workshop on Control and Functions of Two-Dimensional Materials**, 2016 年 6 月 6 日-6 月 6 日, Osaka, Japan（招待）.

103. Kenji Matsuda, "Control over Fluorescence Properties of 1-Cyano-1,2-diphenylethene Derivatives by Structural Modification and Confinement", **The 2nd International Symposium on Aggregation-Induced Emission**, 2015 年 5 月 15 日-5 月 18 日, Guangzhou, China（招待）.

104. Kenji Matsuda, "Assembly and Alignment of Photochromic and Fluorescent Dyes in Two- and Three-Dimensional Way", **The 7th East Asia Symposium on Functional Dyes and Advanced Materials**, 2015 年 9 月 2 日-9 月 5 日, Sakai, Japan（特別）.

105. Kenji Matsuda, "Dynamic Supramolecular Assembly Made of Photochromic Diarylethene at 2D and 3D", **International Symposium for Photo- and Electro-Molecular Machines, PEM2**, 2015 年 10 月 6 日-10 月 7 日, Toulouse, France（招待）.

106. Kenji Matsuda, "Dynamic Supramolecular Assembly Made of Photochromic Compounds at 2D and 3D", **International Symposium on Construction and Application of Functional Molecules/Systems**, 2015 年 10 月 19 日-10 月 21 日, Yilan, Taiwan（招待）.

107. Kenji Matsuda, "Behavior of Supramolecular Assembly Made of Photochromic Diarylethenes at two- and three-Dimension", **10th Phenics International Network Symposium and French-German Winter School**, 2015 年 11 月 23 日-11 月 26 日, Cargèse, France（招待）.

108. Kenji Matsuda, "Photoresponsive Behavior of Photochromic Compounds Self-Assembled in Two- and Three-Dimensional Structures", **CINAM, Aix-Marseille University**, 2015 年 11 月 27 日, Marseille, France（招待）.

109. Kenji Matsuda, "Control over Photophysical and Photochemical Properties by Structural Modification and Confinement", **PPSM, École normale supérieure de Cachan**, 2015 年 11 月 30 日, Cachan, France （招待）.

110. Kenji Matsuda, "Evaluation of beta value of pi-conjugated molecular wires by probing exchange interaction between two nitroxide radicals", **Pacifichem 2015 Syposium#309, Nitroxide Radicals: Synthesis and Functional Bio-/Nanomaterials**, 2015 年 12 月 15 日-12 月 20 日, Honolulu, USA（招待）.

111. Kenji Matsuda, "Photocontrol of Self-Assembly in Solution and at Surface by Photochromism", **Sweden-Kyoto Symposium**, 2014 年 9 月 11 日-9 月 12 日, Stockholm, Sweden（招待）.

112. Kenji Matsuda, "From Color Recognition to Molecular Computers: The Field of Photoresponsive Molecules", **The 11th Japanese-German Frontiers of Science Symposium**, 2014 年 10 月 30 日-11 月 2 日, Bremen, Germany（招待）.

113. Kenji Matsuda, "Photocontrol of 2-Dimensional Self-Assembly of Photochromic Diarylethene at the Liquid/Solid Interface", **The 8th Taiwan-Japan Bilateral Symposium on Architechture of Functional Organic Molecules**, 2014 年 11 月 27 日-11 月 29 日, Fukuoka, Japan（招待）.

114. Tetsuya Nakagawa, "A Photo- and Thermoresponsive Fluorescent Diarylethene Derivative Having Donor-Acceptor Units", **INTERNATIONAL SYMPOSIUM ON PURE & APPLIED CHEMISTRY (ISPAC) 2017** , 2017 年 6 月 8 日-6 月 10 日, Ho Chi Minh, Vietnam（招待）.

115. Yasushi Yokoyama, "Reversible and All-Optical Fine-Tuning of Absorption Wavelength of Photochromic Diarylethene by Photo-Acid-Generating Agent", **Energy Materials Nanotechnology Meeting on Optoelectronics 2016**, 2016 年 4 月 12 日-4 月 15 日, Phuket, Thailand（招待）.

116. Yasushi Yokoyama, "Photosynergetic Regulation of the Photochromic Properties of Diarylethenes", **8th International Symposium on Photochromism (ISOP2016)**, 2016 年 11 月 4 日-11 月 7 日, Shanghai, China（招待）.

117. Takashi Ubukata, "Photoformation of Surface Relief Using Photopolymerizable Bisanthracene", **12th IUPAC International Conference on Novel Materials and their Synthesis (NMS-XII)**, 2016 年 10 月 14 日-10 月 19 日, Changsha, China（招待）.

118. Tetsuya Nakagawa, "Photoswitching of intramolecular donor-acceptor interaction and invisible modulation of the fluorescence ", **INTERNATIONAL SYMPOSIUM ON PURE & APPLIED CHEMISTRY (ISPAC) 2016** , 2016 年 8 月 15 日-8 月 18 日, Kuching, Malaysia（招待）.

119. Y. Yokoyama, "Photon-working switches involving hydrogen bonds", **The International Chemical Congress of Pacific Basin Society 2015**, 2015 年 12 月 15 日-12 月 20 日, Hawaii, USA（招待）.

120. Jiro Abe, "Development of Two-Photon Stepwise Photochromism", **Workshop Nano-synergetics for Young Researchers on Photoactive materials**, 2018 年 3 月 29 日-3 月 30 日, Lille, France（基調）.

121. Jiro Abe, "Fast photochromic molecules toward realization of photosynergetic effects", **First Workshop on Photo-active Nanomaterials with Cooperative and Synergetic Responses**, 2017 年 3 月 27 日-3 月 28 日, Cachan, France（基調）.

122. Jiro Abe, "A new aspect of the stepwise two-photon photochromic reactions", **The 28th International Conference on Photochemistry (ICP2017)**, 2017 年 7 月 16 日-7 月 21 日, Strasbourg, France（招待）.

123. Jiro Abe, Yoichi Kobayashi, "Real-time dynamic holography of a 3D object with fast photochromic molecules", **The 20th International Symposium on Advanced Display Materials & Devices (ADMD2016)**, 2016 年 10 月 19 日-10 月 21 日, Shanghai, China（招待）.

124. Jiro Abe, "Photosynergetics: Stepwise Two-photon photochromic reactions", **International Symposium on Photochromism 2016 (ISOP2016)**, 2016 年 11 月 4 日-11 月 7 日, Shanghai, China（招待）.

125. Jiro Abe, "Fast photochromic molecules toward realization of photosynergetic effects", **9th Asian Photochemistry Conference (APC2016)**, 2016 年 12 月 4 日-12 月 8 日, Singapore, Singapore（基調）

126. Akira Sakamoto, "Constructions of Time-Resolved Raman Spectrometers with Near-Infrared Excitation and Their Applications to Photoexcited p-Conjugated Molecules", **The Fourth Taiwan International Symposium on Raman Spectroscopy**, 2016 年 6 月 28 日-7 月 1 日, Taipei, Taiwan（招待）.

127. Jiro Abe, "Recent Development of Rapid Color-Fading Photochromic Molecules", **Gordon Research Conference (Artificial Molecular Switches &Motors)**, 2015 年 6 月 7 日-6 月 12 日, Boston, USA（招待）

128. Akira Sakamoto, H. Okajima, N. Tanaka, T. Shinmyozu, "Measurements and analyses of the infrared and Raman spectra of radical cation of [34](1,2,4,5)cyclophane: Observation of electron-molecular vibration interaction by infrared spectroscopy and large amplitude bridged ring vibrations by near-infrared resonance Raman spectroscopy", **The International Chemical Congress of Pacific Basin Societies (PACIFICHEM-2015)**, 2015 年 12 月 15 日-12 月 20 日, Honolulu, USA（招待）.

129. H. Okajima, A. Sakamoto, T. Shinmyozu, "Selective Observation of Large Amplitude Inter-ring Vibrations: Resonance Raman Spectroscopy of Cyclophane Radical Cation", **The Third Taiwan International Symposium on Raman Spectroscopy**, 2015 年 6 月 30 日-7 月 3 日, Nantou, Taiwan（招待）.

130. Jiro Abe, "Recent Developments of Fast Photochromic Molecules", **TSRC meeting "Breaking and Making Bonds with Light"**, 2014 年 6 月 30 日-7 月 4 日, Telluride, USA（招待）.

131. Jiro Abe, "Fundamentals and Applications of Fast Photochromic Materials", **XXV IUPAC Symposium on Photochemistry**, 2014 年 7 月 13 日-7 月 18 日, Bordeaux, France（基調）.

132. Yukihide Ishibashi, Tsuyoshi Asahi, "Femtosecond transient spectroscopic study on aqueous organic nanocolloids", **Workshop for Young Researchers on Photo-active materials with Cooperative and Synergetic Responses**, 2018 年 3 月 29 日-3 月 30 日, Lille, France（招待）.

133. Tsunenobu Onodera, Yoshihiko Takeda, Hidetoshi Oikawa, "Recent Progress on Conjugated Polymer Nanofibers: Hybridization and Nonlinear Optical Response", **14th International Conference on Frontiers of Polymers and Advanced Materials (ICFPAM2016)**, 2016 年 10 月 31 日-11 月 4 日, Daejeon, Korea（招待）.

134. Tsuyoshi Asahi, "Light scattering microspectroscopy of single nanoparticles", **2nd Asian Symposium for Analytical Sciences** , 2015 年 9 月 9 日-9 月 10 日, Fukuoka, Japan（招待）.

135. Tsuyoshi Asahi, "Nanoparticle formation by pulsed-laser fragmentation in liquid: Mechanism and applications", **LAMP2015 (The 7th International Congress on Laser Advanced Materials Processing)** , 2015 年 5 月 26 日-5 月 29 日, Kitakyushu, Japan（招待）.

136. Y. Takeda, R. Sato, M. Ohnuma, K. Oyoshi, T. Onodera, H. Oikawa, "Third-order optical nonlinearity of nanomaterials evaluated with fs pump-probe spectroscopy and spectroscopic

ellipsometry", **AMN-7 (The 7th international conference on Advanced materials and Nanotechnology)**, 2015 年 2 月 4 日-2 月 8 日, Nelson, New Zealand（招待）.

137. Tsunenobu Onodera, Hitoshi Kasai, Hidetoshi Oikawa, "Topochemical Polymerization in Diacetylene Nanocrystals and Their Optical Properties", **KJFP-2015 (The 11th Korea-Japan Symposium on Frontier Photoscience - 2015)**, 2015 年 6 月 26 日-6 月 28 日, Jeju, Korea（招待）.

138. Y. Takeda, R. Sato, T. Onodera, H. Oikawa, "Nonlinear optical response of nanomaterials", **EMN (Energy Materials and nanotechnology) Hong Kong Meeting**, 2015 年 12 月 9 日-12 月 12 日, Kowloon, Hong Kong（招待）.

139. Tsuyoshi Asahi, "Pure aqueous colloids of organic compounds prepared by pulse laser fragmentation", **8th International Symposium on Nanomedicine － Fusion of Nanomedicine and Molecular Science of Photosynthesis － (ISNM2014)**, 2014 年 12 月 4 日-12 月 7 日, Matsuyama, Japan（招待）.

140. Tsunenobu Onodera, Hitoshi Kasai, Hidetoshi Oikawa, "Optoelectronic properties of doped Cu-TCNQ nanocrystals", **ISAOP-14 (The 14th International Symposium on Advanced Organic Photonics)**, 2014 年 11 月 4 日-11 月 5 日, Suita, Japan（招待）.

141. Tsuyoshi Tsujioka, "Selective metal-vapor deposition on photochromic surface", **IUPAC 12th International Conference on Novel Materials and their Synthesis (NMS-XII)**, 2016 年 10 月 15 日-10 月 18 日, Changsha, China（招待）.

142. K. Uchida, "Photocontrol of Superhydrophobicity and Superhydrophilicity on Photoresponsive Surfaces", **Toward an international laboratory between France & Japan on Photochemistry**, 2016 年 4 月 1 日-4 月 1 日, Tokyo, Japan（招待）.

143. K. Uchida, "Photocontrol of the Surface Properties and Biological Application by use of Diarylethenes", **MRP (Multi-Responsive Photochrome) Conference**, 2016 年 4 月 25 日-4 月 28 日, Nantes, France（招待）.

144. K. Uchida, "Fractal Surfaces of Molecular Crystals of a Diarylethene Mimicking Lotus Leaf", **9th Asian and Oceanian Photochemistry Conference (APC2016)**, 2016 年 12 月 4 日-12 月 8 日, Singapore, Singapore（招待）.

145. Tsuyoshi Tsujioka, "Selective Metal Deposition on Organic Surfaces for Organic Electronics/Photonics", **EMN-EAST meeting (Energy, Materials, and Nanotechnology)**, 2015 年 4 月 20 日-4 月 23 日, Beijing, China（招待）.

146. Tsuyoshi Tsujioka, "Novel Metal Patterning Method using Vacuum Evaporation on Organic

Surfaces for Electronics/Photonics Applications", **CC3DMR 2015 (Collaborative Conference on 3D & Materials Reseach)**, 2015 年 6 月 15 日-6 月 19 日, Busan, South Korea（招待）.

147. Tsuyoshi Tsujioka, "Selective Metal Deposition on Photochromic Surfaces", **EAS7 (The 7th East Asia Symposium on Functional Dyes and Advanced Materials)** , 2015 年 9 月 2 日-9 月 4 日, Osaka, Japan（招待）.

148. K. Uchida, "Generation of Topographically Versatile Surface, Photo- and Temperature-induced Reversible Changes of Topography of Photochromic Microcrystalline Surfaces", **The Seventh East Asia Symposium on Functional Dyes and Advanced Materials (EAS7)**, 2015 年 9 月 2 日-9 月 5 日, Sakai, Japan（招待）.

149. Seiya Kobatake, "Photomechanical Actuators of Photochromic Crystals", **7th OCARINA International Symposium in conjunction with Symposium in honor of Distinguished Professor Michael Nobel**, 2016 年 3 月 17 日-3 月 18 日, Osaka, Japan（招待）.

150. Seiya Kobatake, "Photomechanical Actuators for Next Generation Intelligent Materials", **The 2014 OCARINA Annual International Meeting**, 2015 年 3 月 4 日-3 月 5 日, Osaka, Japan（招待）.

151. Daichi Kitagawa, Seiya Kobatake, "Photoreversible Current ON/OFF Switching by Photoinduced Bending of Gold-Coated Diarylethene Crystals (Young Scientist Award Presentation)", **22nd International Conference on the Chemistry of the Organic Solid State (ICCOSS 2015)**, 2015 年 7 月 12 日-7 月 17 日, Niigata, Japan（受賞）.

152. Seiya Kobatake, Daichi Kitagawa, "Photomechanical actuators of photochromic diarylethene crystals", **Pacifichem 2015**, 2015 年 12 月 15 日-12 月 20 日, Hawaii, USA（招待）.

153. Seiya Kobatake, Daichi Kitagawa, "Photoresponsive Molecular Machinery of Photochromic Diarylethene Crystals", **Joint Congress of Asian Crystallization Technology Symposium-2014 (ACTS-2014) and 11th International Workshop on Crystal Growth of Organic Materials (CGOM11)**, 2014 年 6 月 17 日-6 月 20 日, Nara, Japan（招待）.

154. Martin Vacha, "Nanoscale characterization of functional polymers and hybrid organic materials by single-molecule spectroscopy", **Indo-Japan Joint Symposium on Polymeric Materials**, 2017 年 1 月 31 日-2 月 1 日, Trivandrum, India（招待）.

155. Martin Vacha, "Light-Harvesting from Single-Molecule Perspective: Selective Turn-on of Energy Transfer by Localized Plasmon and Role of Aggregation in Photophysics of Polyfluorenes", **Interdisciplinary Conference on Light-Harvesting Phenomena and Related Topics LHP 2017**, 2017 年 3 月 26 日-3 月 30 日, Banz, Germany（招待）.

156. Martin Vacha, "Spectroscopy of single nanocrystals of doped I-III-IV semiconductors and organic-inorganic hybrid nanoparticles ", **28th International Conference on Photochemistry**, 2017 年 7 月 16 日-7 月 21 日, Strasbourg, France（招待）.

157. Shuzo Hirata, "Photofunctional materials using long-lived room-temperature triplet excitons", **Royal Society of Chemistry Symposia on Electronic and Photonic Materials**, 2017 年 11 月 17 日-11 月 17 日, Tsukuba, Japan（招待）.

158. Martin Vacha, "Single-molecule study of conformation-related photophysics in conjugated molecular complexes and organic dye-gold nanoparticle structures", **19th Conference on Dynamical Processes in Excited States of Solids**, 2016 年 7 月 17 日-7 月 22 日, Paris, France（招待）.

159. Martin Vacha, "Nanoscale characterization of functional hybrid organic materials by single-molecule spectroscopy", **2016 International Symposium on Integrated Molecular/Materials Science & Engineering**, 2016 年 10 月 14 日-10 月 15 日, Qingdao, China（招待）.

160. Martin Vacha, "Nanoscale characterization of functional polymers and hybrid organic materials by single-molecule spectroscopy", **Asia Conference on Nanoscience and Nanotechnology AsiaNANO 2016**, 2016 年 10 月 10 日-10 月 13 日, Sapporo, Japan（招待）.

161. Shuzo Hirata, "White afterglow room temperature emission from isolated single aromatic unit", **2016 International Symposium on Integrated Molecular/Materials Science & Engineering**, 2016 年 10 月 14 日-10 月 15 日, Qingdao, China（招待）.

162. Martin Vacha, "Molecular-level study and control of conformation and photophysics in conjugated molecular complexes", **11th International Conference on Optical Probes of Conjugated Polymers and Organic Nanostructures**, 2015 年 6 月 14 日-6 月 19 日, Hong Kong（招待）.

163. Martin Vacha, "Single-molecule study and control of conformation-related photophysics in conjugated molecular complexes", **12th International Conference on Hole Burning, Single Molecule and Related Spectroscopies**, 2015 年 8 月 24 日-8 月 27 日, Tartu, Estonia（招待）.

164. Martin Vacha, "Molecular-level study and control of conformation and photophysics in conjugated molecular complexes", **The International Chemical Congress o Pacific Basin Societies 2015**, 2015 年 12 月 15 日-12 月 20 日, Honolulu, USA（招待）.

165. Shuzo Hirata, "Large Nonlinar Abosrption under Weak Conteneous Incoherent Light using Long-lived Room Tempeature Excitons", **The 32 nd International Conference of Photopolymer**

Science and Technology, 2015 年 6 月 24 日-6 月 26 日, Chiba, Japan（招待）.

166. Martin Vacha, "Single molecule spectroscopy: A window to the nanoworld of polymers and complex soft matter", **International Conference on Smart Systems Engineering 2014**, 2014 年 10 月 15 日-10 月 17 日, Yonezawa, Japan（招待）.

167. Martin Vacha, "Spectroscopy of single conjugated polymer chains: electroluminescence, photoluminescence and mechanical control of photophysics", **8th Asian Photochemistry Conference** , 2014 年 11 月 10 日-11 月 13 日, Trivandrum, India（招待）.

168. Vasudevan Pillai Biju, "Quantum Dots for Single-Molecule Bioimaging: Challenges and Prospects", **International Conference on Nanoscience and Nanotechnology-ICONSAT 2016**, 2016 年 2 月 29 日-3 月 2 日, Pune, India（招待）.

169. Vasudevan Pillai Biju, "Nanoparticles for Bioimaging: Potentials and Pitfalls", **INTERNATIONAL CONFERENCE ON MATERIALS FOR THE MILLENIUM – "MATCON 2016"**, 2016 年 1 月 14 日-1 月 16 日, Cochin, India（招待）.

170. Morihiko Hamada, Shunsuke Nakanishi, Vasudevan Pillai Biju, "Photoluminescence Enhancement and Quenching in Single Semiconductor Quantum Dots", **INTERNATIONAL CONFERENCE ON MATERIALS FOR THE MILLENIUM – "MATCON 2016"**, 2016 年 1 月 14 日-1 月 16 日, Cochin, India（招待）.

171. Vasudevan Pillai Biju, Shin-ichi Yamashita, Morihiko Hamada, Shunsuke Nakanishi, Shin-ichi Wakida, Hironobu Saito, Yoshio Nosaka, "Anti-oxidation of semiconductor quantum dots by Auger ionization revealed by single-molecule fluorescence imaging", **PacifiChem2015**, 2015 年 12 月 15 日-12 月 20 日, Honolulu, USA（招待）.

172. Vasudevan Pillai Biju, "Single Molecule Fluorescence: From Sensing to Sensitization", **International Syomposium on Photonic Applications and Nanomaterials**, 2015 年 10 月 28 日-10 月 30 日, Trivandrum, India（招待）.

173. Vasudevan Pillai Biju, "Single-molecule Detection using Quantum Dot Labels: Challenges and Prospects", **HOKUDAI-NCTU Joint Symposium on Nano, Photo and Bio Sciences in 2016**, 2016 年 10 月 4 日-10 月 5 日, Sapporo, Japan（招待）.

174. Vasudevan Pillai Biju, "Blinking and Bleaching Quantum Dots in Single-molecule Detections", **Asian Photochemistry Conference 2016**, 2016 年 12 月 4 日-12 月 8 日, Singapore, Singapore（招待）.

175. Vasudevan Pillai Biju, "Luminescent Semiconductor Nanocrystals: Challenges and Prospects",

International Symposium on “New Trends in Applied Chemistry" (NTAC2017), 2017年2月9日-2月11日, Kochi, India（招待）.

176. Vasudevan Pillai Biju, "Luminescent Quantum Dots in Biomaging", **The 17th RIES-HOKUDAI INTERNATIONAL SYMPOSIUM on 柔**, 2016年12月13日-12月14日, Sapporo, Japan（招待）.

177. Vasudevan Pillai Biju, "Incessant and Intermittent Photoluminescence of Quantum Dots in Single Molecule Detections", **AsiaNANO2016** , 2016年10月10日-10月13日, Sapporo, Japan（招待）.

178. Vasudevan Pillai Biju, "A Few Intracellular Pathways Of Quantum Dot Bioconjugates", **MAF2017**, 2017年9月10日-9月13日, Bruges, Belgium（招待）.

179. Vasudevan Pillai Biju, "Single molecule detections & Bioimaging using Semiconductor Nanocrystals", **Emerging Frontiers in Chemical Science 2017** , 2017年9月23日-9月25日, Calicut, India（招待）.

180. Vasudevan Pillai Biju, "Quantum Dots for Single Molecule Detections: Recent Progress and Challenges", **2017 RIES-CIS Symposium** , 2017年10月27日-10月28日, Hsinchu, Taiwan（招待）.

181. Vasudevan Pillai Biju, "Semiconductor Quantum Dots in Single Molecule Detections: Prospects and challenges", **ICPA**, 2017年11月10日-11月13日, Kottayam, India（招待）.

182. Vasudevan Pillai Biju, "Semiconductore Quantum Dots for Single-molecule Detections in Living Cells: Challenges and Progress", **Nanobiotechnology**, 2017年12月6日-12月6日, Trivandrum , India（招待）.

183. Vasudevan Pillai Biju, "Semiconductor Quontum Dots in Bioimagion: Progress and Challenges", **9th Bangalore India NANO** , 2017年12月7日-12月9日, Bangalore, India（招待）.

184. Hiroshi Uji-i, "Plasmonic endoscopy toward single cell interrogation", **RACI National Centenary Conference 2017**, 2017年7月23日-7月28日, Melbourne, Australia（招待）.

185. Hiroshi Uji-i, "Plasmonic Waveguiding Remote Excitation Spectroscopy toward Single-cell Interrogation", **15th Conference on Methods and Applications in Fluorescence**, 2017年9月10日-9月13日, Bruges, Belgium（招待）.

186. Hiroshi Uji-i, "Remote Spectroscopy Using Plasmonic Waveguide toward Single-Cell Interrogation", **2017 RIIES-CIS Symposium**, 2017年10月27日-10月28日, Hsinchu, Taiwan（招待）.

187. Kosei Ueno, "Chemical Applications of Plasmonics Using precisely Controlled Metallic Nanostructures", **The 7th International Conference on Surface Plasmon Photonics (SPP7)**, 2015 年 5 月 30 日-6 月 3 日, Jerusalem, Israel（招待）.

188. Kosei Ueno, "Spectral Properties and Dynamics of Plasmon Nanostructures", **XVIIIth International Krutyn Summer School 2015**, 2015 年 9 月 27 日-10 月 3 日, Krutyn, Poland（招待）.

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191. Kosei Ueno, "Surface-Enhanced Terahertz Spectroscopy Using Plasmonic Structures", **CityU/Hokkaido University Joint Workshop "Advanced Materials and Characterisation"**, 2015 年 12 月 10 日-12 月 11 日, Hong Kong, Hong Kong（招待）.

192. Kosei Ueno, "Fabrication and Plasmonic Application of Nano-Engineered Gold Particles", **International Conference on Materials for the Millennium (MATCON2016)**, 2016 年 1 月 14 日-1 月 16 日, Cochin, India（基調）.

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194. Kosei Ueno, "Plasmon-Based Physics and Chemistry Using Nano-Engineered Gold Nanoparticles", **The 11th Annual IEEE International Conference on Nano/Micro Engineered and Molecular Systems (IEEE-NEMS 2016)**, 2016 年 4 月 17 日-4 月 20 日, Miyagi, Japan（招待）.

195. Kosei Ueno, "Plasmonic-Enhanced Photochemical Reactions Using Metallic Nanoparticles", **The 10th Asia-Pacific Laser Symposium (APLS2016)**, 2016 年 5 月 10 日-5 月 14 日, Jeju Island, Korea（招待）.

196. Kosei Ueno, Tomoya Oshikiri, Hiroaki Misawa, "Plasmon-Induced Artificial Photosynthesis; Evolution of Hydrogen and Ammonia", **The 6th Sino-Japan bilateral young chemist forum**, 2016 年 6 月 30 日-7 月 2 日, Dalian, China（招待）.

197. Kosei Ueno, "Infrared and THz Plasmonics Using Nano-Engineered Gold Structures", **7th International Conference on Metamaterials, Photonic Crystals and Plasmonics (META 2016)**, 2016 年 7 月 25 日-7 月 28 日, Torremolinos (Malaga), Spain（招待）.

198. Kosei Ueno, "Spectral Modulation of Molecular/Intermolecular Vibrational Mode by Infrared Plasmon", **Progress in Electromagnetics Research Symposium 2016 (PIERS 2016)**, 2016 年 8 月 8 日-8 月 11 日, Shanghai, China（招待）.

199. Kosei Ueno, Hiroaki Misawa, "Surface Plasmon-Assisted Chemical Reactions Using Nano-Engineered Gold Nanoparticles", **The 14th International Conference of Near-Field Optics (NFO-14)**, 2016 年 9 月 4 日-9 月 8 日, Hamamatsu, Japan（招待）.

200. Kosei Ueno, Tomoya Oshikiri, Hiroaki Misawa, "Plasmon-induced water splitting by visible and near infrared light using gold nanostructured oxide semiconductors", **Global Artificial Photosynthesis - Breakthroughs for the Sustainocene**, 2016 年 9 月 9 日-9 月 11 日, Lord Howe Island, Australia（招待）.

201. Kosei Ueno, Sho Nozawa, Jie Li, Quan Sun, Tomoya Oshikiri, Hiroaki Misawa, "Spectral Modulations Induced by Interactions between Molecular Vibrational Modes and Infrared Plasmon; Strong Coupling or Electromagnetically Induced Transparency", **The 8th International Workshop on Advanced Materials Science and Nanotechnology (IWAMSN 2016)**, 2016 年 11 月 8 日-11 月 12 日, Ha Long City, Vietnam（招待）.

202. Kosei Ueno, Sho Nozawa, Jie Li, Quan Sun, Tomoya Oshikiri, Hiroaki Misawa, "Infrared Plasmonic Chemistry Based on Strong Coupling between Gold Nanostructures and Molecular/Intermolecular Vibrational Modes", **9th Asian and Oceanian Photochemistry Conference (APC2016)**, 2016 年 12 月 4 日-12 月 8 日, Singapore, Singapore（招待）.

203. K. Ueno, S. Nozawa, H. Yu, J. Li, Q. Sun, T. Oshikiri, H.Misawa, "Spectral Properties of Plasmon-Molecule Hybrid States and Coupled Plasmonic Systems", **The 8th RSC‐CSJ Joint Symposium —Recent Developments in Plasmonics— The Chemical Society of Japan The 97th Annual Meeting**, 2017 年 3 月 18 日-3 月 18 日, Yokohama, Japan（招待）.

204. Kosei Ueno, Quan Sun, Hiroaki Misawa, "Near-field spectral properties of coupled plasmonic systems", **Progress In Electromagnetics Research Symposium 2017 (PIERS 2017)**, 2017年5月 22 日-5 月 25 日, St. Petersburg, Russia（招待）.

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Their Shape Anisotropy", **231st ECS Meeting**, 2017 年 5 月 28 日-6 月 1 日, New Orleans, USA（招待）.

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267. Yutaka Wakayama, "Molecules meet Si: bridging single-molecular function with practical device", **Trends in Nanotechnology International Conference 2015**, 2015年9月7日-9月11日, Toulouse, France（招待）.

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271. Yoshiyuki Kageyama, "Self-organization of molecular machine for autonomous macroscopic motion under non-equilibrium steady state", **The First MIRAI Seminar**, 2017年10月17日-10月19日, Lund, Sweden（招待）.

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291. Keiko Tawa, "Sensitive Fluorescence Imaging of Cells with a Palsmonic Chip", **2015 KJF International Conference on Organic Materials for Electronics and Photonics**, 2015 年 9 月 6 日-9 月 9 日, Jeju island, Korea（招待）.

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295. Takahiro Seki. Daichi Ymaoka, Mitsuo Hara, Shusaku Nagano, "Photoalignable Radical Initiator for Anisotropic Polymerization in Liquid Crystalline Media", **19th International Symposium on Advanced Display Materials and Devices (ADMD 2015)**, 2015 年 9 月 22 日-9 月 23 日, Gyeongju, Korea（招待）.

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298. Takahiro Seki, Shusaku Nagano, "Free surface command layer for the photoalignment", **KJF International Conference on Organic Materials for Electronics and Photonics 2016 (KJF-ICOMEP)**, 2016 年 9 月 4 日-9 月 7 日, Fukuoka, Japan（招待）.

299. Takahiro Seki, "Free surface command layer for the photoalignment of polymer liquid crystals", **Rad Tech Asia 2016**, 2016 年 10 月 24 日-10 月 27 日, Tokyo, Japan（招待）.

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301. Takahiro Seki, "New stragegies for t he photoalignment of liquid crystalline polymers", **The 11th SPSJ International Polymer Conference (IPC2016)**, 2016 年 12 月 13 日-12 月 16 日, Fukuoka, Japan（招待）.

学会発表総数

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以下の表には、研究班ごとの発表数を示す。公募班については 27－30 年度としてまとめている。共同研究発表による重複は除いていない。これらの数字には、招待講演なども含まれている。

計画班（平成 26－30 年度）

	国内	国際
宮坂	237	165
井村	84	30
重田	159	102
玉井	145	73
河合	335	98
前田	223	130
松田	155	65
横山	153	72
阿部	142	102
朝日	156	51
内田	141	57
小畠	215	42
Vacha	43	23
小計	**2188**	**1010**

公募班　（平成 27－30 年度）

	国内	国際
上野	25	28
鳥本	56	32
磯崎	10	7
久保	61	36
柳井	3	5
Biju	6	35
雲林院	7	3
岡	0	2
坂本	4	3
深港	34	18
佐田	68	13
山村	32	12
齊藤	91	14
森	79	29
羽曽部	71	7
若山	0	10
宍戸	51	15
佐藤	126	57
則包	13	10
梶本	7	1
伊藤	38	19
関	112	40
池田	59	44
森本	71	12
田和	50	34
景山	12	5
小野寺	20	10
出羽	42	10
小計	**1148**	**511**

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研究成果

電子励起状態分子はエネルギーや物質変換、光機能発現等において重要な役割を果たしている。これら励起分子の諸過程を対象とする光化学関連研究分野において、日本の研究者は国際的にも先導的役割を果たし、励起分子素過程、光物質合成、光機能分子、光触媒、太陽電池、光物質プロセッシングなど、広範な分野の発展に多大な貢献を行ってきた。

しかし光機能発現やエネルギー・物質変換に応用が想定される比較的大きな分子では、図 1 に示すように、

① 高位電子状態が生成しても迅速に最低励起状態に緩和し（Kasha 則）（光量子の質の損失）

② 集合体系において多くの励起分子が生成しても分子間の迅速な消滅過程（励起子 annihilation）により最終的にはごく少数の励起分子しか残らない（光量子の量の損失） など、光子の質や量の損失に関わる大きな制限が存在する。更に、

③ 通常の光吸収で到達できる電子状態は主に 1 光子許容なものに限られており、分子が本来有する多様な電子励起状態を有効に利用することも困難であった（多様性の損失）。

これらの制限のために、分子・物質系の電子状態選択的な応答や多数の励起分子やその応答が協調して作動する光機能発現、これらの結果として進行するメゾスコピック・マクロスコピックスケールの分子集合体系の光応答・機能などへの高次展開は困難なものとされ、主な研究対象は“1 光子吸収による最低励起状態からの 1 分子の光応答” に限られてきた。

しかし近年、本提案領域の代表者や参画者の研究を含め、多光子吸収や多重励起を利用した高位・禁制電子状態における選択的・特異的光反応、高位電子励起状態からの複数励起子の生成、局所場等の利用による摂動を超えた新奇電子状態作成、複数光子によって誘起される多重スイッチ過程、また集合体系における多数励起分子の協同的・階層的光力学応答など、これら 3 種の制限を超える新現象が報告されだしていた。

これらの背景を基に、多重・多光子励起、電子状態変調、分子・集合体開発 等の方法を用い、従来の “1 光子吸収と 1 分子応答” を超えた光子有効利用を可能とす複合光応答（英語では Photosynergetics と標記した）系の開拓・発展により、短期的な光化学分野における国際的優位性の継続のみならず、中長期的にも今後の解決すべき分子系の光利用関連諸課題の共通基盤となる学理の構築を目標とした。

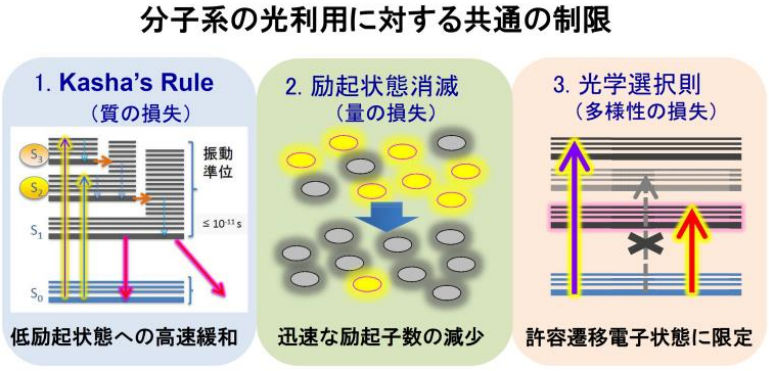

図 1．凝縮系の電子励起状態分子に対する制限

研究の推進のために、本新学術領域では、A01 から A03 までの３つの研究領域を組織した（図 2）。

図 2. 組織構成

A01 班は**３種の制限に対する正面からのアプローチ**を目的として、多重、多光子励起による高位電子状態からの反応開拓・機構解明、局所場を利用した電子状態変調、光反応、励起子分裂・融合等を課題として、実験物理化学、計算化学、分子や物質開発などの領域の研究を中心として、本研究推進の基礎となる複合励起による素過程制御と新規光応答・反応開拓に関する研究を行った。
［計画班 4，公募班 6（H27-28）、7（H29-30）］

A02 班では、annhilation **などによる“量の損失”を超えるための多数分子が協調する複合反応ネットワークや反応場の構築**を目的とした。分子・分子集合体の設計と合成手法を用い、１分子レベルの機能からメゾスコピック・マクロレベルの物性変化への展開に必要となる“加算性”や“増幅性”が確保できる光機能システムや反応網の開拓・確立を課題として、分子・分集合体の合成・開発、新規物性開拓を中心とした研究者が、分子開発、界面分子配列、集積化、反応場構築などの観点から研究を行った。
［計画班 4，公募班 7（H27-28）、7（H29-30）］

A03 班では、比較的大きなサイズ（数 10 nm から数 μm 以上）において**多数の光子や多数の励起分子による反応がメゾ、マクロ複合応答へとつながる光応答系の開拓、機構解明、高度展開**を主目標とした。集合系に特徴的な協奏的、協同的な複合過程を利用し、複合励起と応答を顕在化できる光駆動する分子集合系、多光子吸収により駆動する光応答系や、光強度に閾値を持つ光応答系など、１光子１分子光反応では実現できない高度な集合体構造変化に基づく複合光応答システム創出を課題とし、分子設計・開発、集合体構築、固体化学、材料化学、固体物性測定などの研究者を中心に、主に集合体の応答に基軸を置いた研究を行った。
［計画班 5，公募班 6（H27-28）、7（H29-30）］

総括班はこれらの研究の推進にあたり共同研究提案や実施支援を行う（約 200 件）と共に、若手育成についても積極的に活動をった。また国際活動支援班は国際共同研究の支援（約 100 件）のみならず、フランスとの間では CNRS プログラムとして「日仏光化学共同研究所」を設置し中長期的国際共同研究体制を構築した。

以下に代表的な成果を A01、A02、A03 班に分けて示す。これらの中には、本来の課題の研究の進捗・発展の中で、たまたま見出されたものもある。そのため、個々の研究成果としては研究者の所属の班ではなく、その内容に従い A01、A02、A03 に分類して示した。また、ここに示した結果には、領域内、また国際共同研究として行われたものも多い、このような場合にも、その内容に従い分類した。

A01 班： **特異電子状態へのアプローチ**

高位電子励起状態の反応機構解明と新規反応開拓、局所場による電子状態変調と特異電子状態へのアクセス手法の開発、多重励起子状態の制御応用と励起子融合と分裂等を用いた新規光応答の開拓を目的とし、計画班・公募班が、A02、A03 との共同研究を含めて、研究を展開した。

高位電子励起状態からの反応機構解明・開拓

超短寿命（<<10^{-12} 秒）で最低電子励起状態へと迅速に緩和する高位電子励起状態からの反応を支配する因子の解明、新規反応開拓は、Kasha 則の超克に対する重要な課題である。

フルギドやジアリールエテン誘導体など 6π 電子系の光誘起環開閉異性化反応では、可視逐次２光子励起による閉環体の高位励起状態から高効率に開環反応が進行する。一方、同程度のエネルギーに対応する紫外１光子励起では、このような高効率開環反応は進行しない。宮坂グループ (G) では、自らが見出したこの電子状態選択的な高位励起状態反応を支配する因子の解明のために、フェムト秒 2 パルス励起による反応ダイナミクスの高精度測定システムを構築し、これを機構解明に応用した。その結果、１光子励起直後の 1B 状態から光吸収により生成した高位励起状態からは開環反応が進行しないが、2A 状態から生成した高位励起状態からは高収率で開環反応が進行すること、また 2A 状態における構造緩和後にもう１光子吸収した場合には、より高収率に開環反応が進行することを明らかにした（図 3）。また非共鳴同時２光子吸収の結果とも比較し、高位電子状態の開環反応も基本的には電子波動関数の対称性に基づき進行すること、また非常に短い（<<1ps）高位励起状態寿命間には大きな構造変化が起こらないため、2 光子目の吸収の始状態の核配置が反応に大きな役割を果たすことを示し（*JACS* 2017、*JPCL* 2017、*JPCC* 2018 *Suppl. Cover*, *PCCP* 2018 *Back cover*, A03 小畠 G、A02 横山 G との共同研究）、この振動位相に同期した 2 パルス反応制御を実現した。

また逐次２光子励起を光イオン化の機構解明に応用し、低エネルギーで進行する溶液中の電子放出では、サブピコ秒程度の寿命を持つ特異電子状態から電子放出が進行することを明らかにした（*PCCP* 2019 *Back Cover*）。さらに A03 阿部 G との共同研究により、可視逐次２光子吸収で生成した高位励起状態からの選択的電荷分離反応を経由したラジカル解離（*JACS* 2017）半導体量子ドットの高励起状態からの選択的電子移動反応（*JPCL* 2018 *Suppl. Cover*）など、新規２光子誘起電子移動系を開拓した。

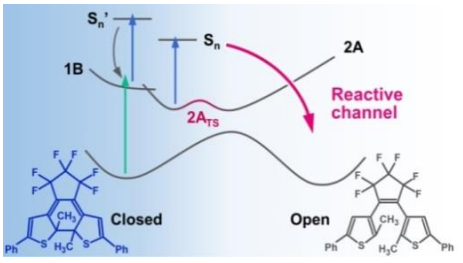

図 3. 逐次2光子開環反応の機構.

玉井 G では半導体ナノ粒子を対象に、高位励起状態か

らの超高速ホット電子移動系を構築し、半導体ナノ粒子においても高位電子励起状態の有効利用を実証した(*JPCC* 2016)。

柳井 G は大分子系の高位電子励起状態の計算手法を開発し(*Nature* 2016, *J. Chem. Theory Comput.* 2017 *Editors' choice*)、実験結果と対応させることによりその有効性を検証した(*JACS* 2017, 阿部 G、宮坂 G との共同研究)。この結果、理論化学による大分子の高位励起状態の詳細な先導的研究が可能となった。

また、阿部 G では、多重励起の応用として、入力光強度に閾値を有する非線形光応答の開拓を目的に、フェノキシルラジカルとイミダゾリルラジカルを基盤とする新規高速フォトクロミック複合体(PIC)を開発し(図 5、*JACS* 2015)、分子系の複合化を行うことにより照射光強度に依存する着色状態の色調変化に成功し、着脱色の2値応答から多値応答系へと発展させた(*JACS* 2018, *Suppl. Cover*)。また、二つの PIC がそれぞれベンゼン環のパラ位に結合した bisPIC は、1光子反応では短寿命の着色体を、一方2光子反応では長寿命の着色体をそれぞれ生成することを示し(*JACS* 2017)、光強度に依存する分子系への応用展開に必用な分子設計指針を示した。

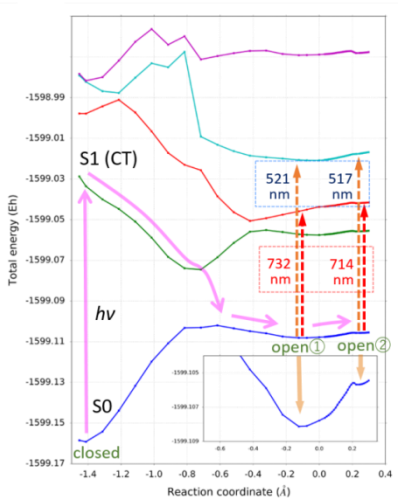

図４．高位電子状態に対応可能な高精度計算手法の開発と応用.

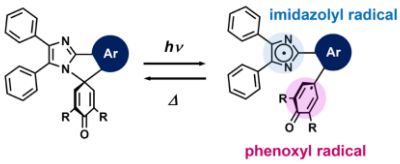

図 5. PIC の多重光反応.

多重励起子の利用

迅速に進行する励起子消滅を防ぎ有効に利用することは“量の損失”に対する重要な課題である。三重項消滅(TTA)に基づくアップコンバージョン(UC)や励起子分裂(Singlet Fission: SF)は、この課題に密接に関連すると共に分子レベルの光応答を増幅しメゾスコピック複合光応答システムにつなぐための重要な素過程である。

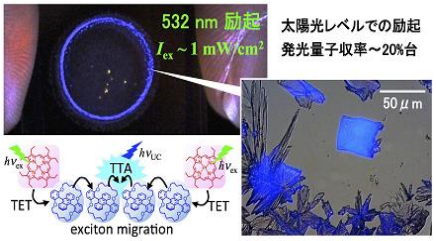

図 6. 高効率 UC 固体系の構築.

重田 G は理論と実験の協奏により研究を展開し、詳細な機構解明を行い(*JPCC* 2018, *JPCL* 2018)、これに基づき高効率 UC 結晶性固体を実現した(図 6、*Mater. Horiz.* 2017, A03 Vacha G との共同研究)。

Biju G は高発光性 QD における多励起子による発光 blinking の抑制機構を明らかにし、多励起子の有効利用法を開発した(*ACIE* 2019, Hot Article, Cover Page)。

坂本 G は多重励起子のアップコンバージョンを利用し赤外光により駆動する高効率水素生成光ナノ粒子触媒を開発した(*JACS* 2019, *Suppl. Cover*, 玉井 G との共同研究)。

局所場による電子状態変調

禁制電子状態を含む電子励起状態の顕在化、また、局所場の利用による摂動を超えた新たな電子状態の作成は“多様性の損失”を超え、新たな励起状態の応答の開拓を可能とする。

井村 G では、貴金属ナノ構造近傍の増強電場による分子系の電子状態変調を対象に、メソ光増強場の種々の可視化法を開発・応用し、二次元プラズモンの空間構造と光励起状態の長寿命化、

さらにメソ構造体の近接場光選択則を確立し（図 7、*Nano Lett.* 2015, *JPCL* 2018）、局所場を利用した電子励起状態変調に対する基礎的原理を構築した。

井村 G では、貴金属ナノ構造近傍の増強電場による分子系の電子状態変調を対象に、メソ光増強場の種々の可視化法を開発・応用し、二次元プラズモンの空間構造と光励起状態の長寿命化、さらにメソ構造体の近接場光選択則を確立し（図 7、*Nano Lett.* 2015, *JPCL* 2018）、局所場を利用した電子励起状態変調に対する基礎的原理を構築した。

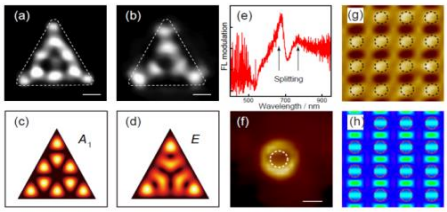

図7. メソ光増強場の可視化法の開発と応用.

上野 G は、励起子ープラズモン強結合による電子状態変調を目的として、振動モード強結合による無輻射失活速度定数の変調を実証する（*Nature Nanotechnol.* 2018）とともに光反応制御やプラズモン状態の制御も可能とした（*Nature Com.* 2018）。

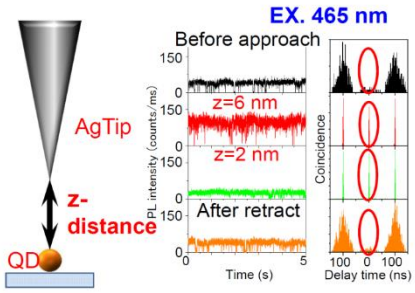

図 8. 半導体 QD の励起子消滅過程の制御.

増尾 G（玉井 G 分担者）は、半導体量子ドット周辺の空間の増強場を精密に制御することで励起子消滅を回避可能であることを実験的に示した（図 8, *ACS Photonics* 2016）。

また、坂本らは、ナノ粒子の局在化プラズン共鳴励起による熱ホールの移動プロセスを初めて明らかにし、プラズモン誘起熱キャリア移動の機構の中でも高い量子収率と、圧倒的に長い電荷分離状態（従来の 10 万倍）を有する材料を開発した（*Nature Com.* 2018）。

以上のように、実験および理論のアプローチにより、また他領域との共同研究により高位励起状態の反応機構解明と新規反応開拓、また、多重励起子状態の制御応用と励起子融合と分裂等を用いた新規光応答、局所場による電子状態変調と特異電子状態へのアクセス手法の開発と応用など、目的に沿った研究成果が得られた。

A02 班：多分子協調場と手法の開拓

Annhilation などによる“量の損失”を超えるための多分子協調による加算・増幅的複合応答系の開拓、また、個々の分子の光応答を集合体複合応答へと連結する「つなぐ仕組み」の開発を目標に、材料化学、合成化学、物理化学、理論化学などを中心とする計画班・公募班が、A01 や A03 との積極的な共同研究を含め、多数分子が協調する複合反応ネットワークや反応場の構築・開発を中心に研究を展開した。

<u>増幅光応答系の開拓</u>

加算・増幅的な反応系の作成は、上記の通り、“量の損失”を超えるためのアプローチであるだけでなく個々の分子の光応答を集合体複合応答へと連結するための重要な課題である。

河合 G では、光のみならず酸化により異性化反応を行うターアリーレン誘導体を見出し、連鎖酸化異性化反応により実効的な収率として 100,000 %の反応増幅ネットワークを構築し、光誘起電子移動をトリガーとした場合 3,000%以上の光反応量子収率が得られることを実証した（図 9, *Chem.*

Eur. J. 2016, *Inside Cover*)。さらに、連鎖反応を誘起する光反応系として最高感度（反応量子収率 71%）の光酸発生剤の開発、光塩基発生剤への展開、また光ルイス酸発生剤の開発も行い、1000%を超える実効反応量子収率で、典型的なルイス酸触媒反応例の向山・アルドール反応の光駆動に成功したするとともに（*Chem. Com.* 2017, *JACS* 2015, *JACS Spot Light,*阿部 G との共同研究）、光塩基発生剤の開発へも研究を展開し（*JOC* 2018）、多様な光反応の増幅方法を開拓した。

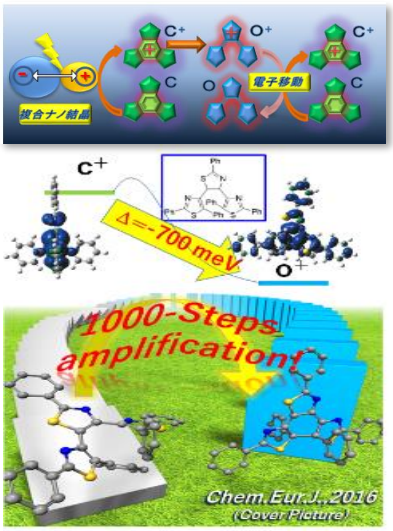

図 9. 超増幅性を有する異性化反応.

深港 G では蛍光性ユニットと光応答ユニットを連結した複合光応答分子の自己集積により蛍光性ナノ粒子を作製し、光異性化反応により生成した構成分子のわずか数%の異性体が、効率的な励起エネルギー受容体として作用することで、ナノ粒子全体の蛍光を完全に ON→OFF スイッチングできることを示し、発光制御における“１光子－１分子応答”から“少数光子－多分子応答”への増幅光応答系を構築した（図 10、*ACIE* 2016, *Cover Picture*）。

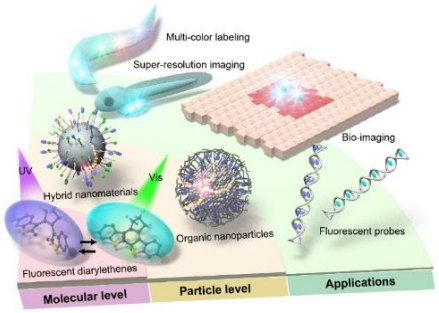

図 10. 分子ナノ粒子における“少数光子－多分子応答”への増幅光応答系.

羽曾部 G は、A01 とも深く関連する有機分子の多励起子生成反応である一重項分裂とそのエネルギー変換過程の高効率化を目標に、アセン系分子の二量体から有機・無機複合材料の合成と光物性評価に取り組んだ。特に顕著な成果としてペンタセンおよびテトラセン二量体における量論的な一重項分裂の反応進行（三重項量子収率～200%）とその長寿命化を実現した。さらにクロラニルを電子受容体として利用することで、約 170%の電子移動量子収率を達成した（図 11、*ACIE* 2019, *ACS Energy Lett.* 2019）。

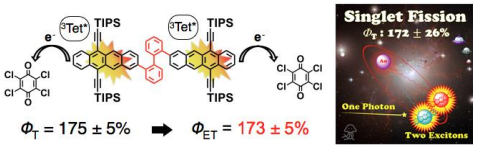

図 11.　高効率一重項分裂電荷分離系.

佐藤 G では凝集分子系における励起状態の損失抑制に関する理論構築を行い、内部転換による励起状態緩和の抑制をもたらす因子として複数の分子の高対称性偽オリゴマー形成に起因する電子状態の擬縮退の重要性を示し、増幅応答系構築に必要な集合体設計指針を示した(*Sci. Rep.* 2017)。

横山 G では、タンパク質をテンプレートとすることでアキラルな光異性化反応の高エナンチオ選択的完全キラル制御を実現（*Chem.Com.* 2017）し、しキラリティーの光増幅手法を示した。また、フォトクロミック特性のオンオフを光のみによって制御する光―光制御系にも研究を展開し（*Org. Lett.*2016）、これら光増幅系の複合化に必用な手法を開拓した。

<u>集合化に伴う多機能光応答分子集合体の設計・開発</u>

集合体における光反応の加算・増幅的な応答は、A03 にも関連する課題である。特にボトムアップ的なアプローチは、個々の分子の光応答を集合体複合応答へと連結するための重要な合理的

指針を与える。

松田 G ではジアリールエテンを骨格とする両親媒性光応答分子を合成し、下部臨界完溶挙動と組み合わせ、光によるマクロな可逆形態制御を実現した（*JACS* 2015）。さらに光異性化分子の固液界面自己組織化における多重光機能の解明に取り組み、液中 STM による分子像観察にもとづく表面被覆率の濃度依存性から光異性化反応による光自己組織化の配列形成メカニズムを解明するとともに、非線形光応答性を示す自己組織構造形成へと発展させた（図 12、*ACIE* 2017）。

図 12. 二次元自己組織化における協働的光応答系.

斎藤 G は基底状態では非平面構造を持つ 8π 電子系シクロオクタテトラエン型分子が、励起状態では平面構造へと大きな構造変化を行うことを見出し、分子レベルの構造変化に基づく光誘起融解性集合体を構築し、多数光子の照射により剥離可能な接着集合体として機能することを示した（図 13、*Nature Com*. 2016）。更にその励起状態挙動を解明すると共に（*J. Mater. Chem. C*, 2017, 宮坂 G との共同研究）、アントラセンの光 2 量化を組み合わせた光融解性液晶接着剤の開発、基底状態構造の強制的平面化によるメカノフォア機能の発現など、多分子の協同的応答による新規光応答系を実現した。

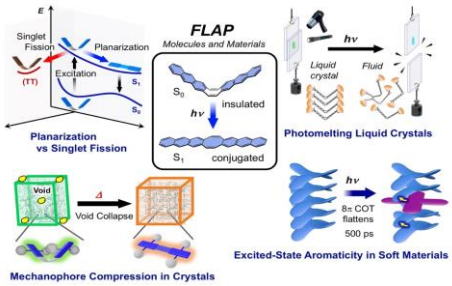

図 13. シクロオクタテトラエン型分子の多重光機能.

前田 G では、アゾベンゼンをアニオンユニットとしてイオン結晶中に導入することで光異性化に伴う可逆的な結晶–結晶相転移に成功した（,図 14、*Chem. Eur. J.*, 2016 & 2017）。

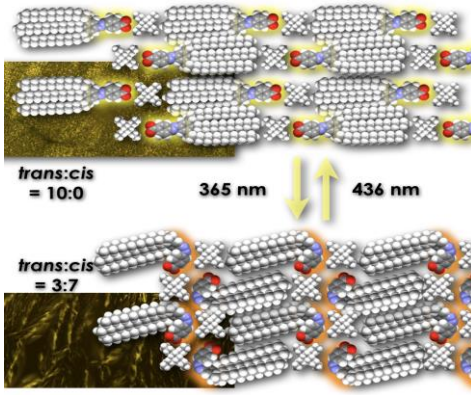

図 14. 光異性化誘起相転移結晶の作動原理.

またこれらの集合系の多機能光応答に必要な構成分子の配向制御手法として、宍戸 G では光照射の位置や強度変化により簡便かつ高精度に大面積の分子配向パターンを形成できる光重合手法を開発し、光メカニカル挙動の制御に応用した（*Sci.Adv.* 2017）。

以上のように、分子協調による加算・増幅的複合応答系の構築に関わる増幅反応ネットワーク、増幅型多機能光応答分子集合体の構築、機構解明が多数行われた。

A03 班：**高次光応答分子システムの構築**

比較的大きなサイズ（数 10 nm から数 μm 以上）において多数の光子や多数の励起分子による反応がメゾ、マクロ複合応答へとつながる光応答系の開拓、機構解明、高度展開を主目標とした。集合系に特徴的な協奏的、協同的な複合過程を利用し、複合励起と応答を顕在化できる光駆動する分子集合系、多光子吸収により駆動する光応答系や、光強度に閾値を持つ光応答系など、１光子

１分子光反応では実現できない高度な集合体構造変化に基づく複合光応答システム創出を課題とし、分子設計・開発、集合体構築、固体化学、材料化学、固体物性測定などの研究者を中心に、主に集合体の応答に基軸を置いた研究を行った。

光駆動する集合応答系の機構解明と機能開拓

集合化された光異性化分子系において、分子レベルの光反応が協同的な結晶変形などのマクロスコピックな光応答を誘起することは、既に A03 への参画研究者の中から見出されていた。これらの現象の機構解明、分子レベルを超えた集合体としての支配因子抽出、新規応答の開拓は、複合光応答の領域全体の展開に対しても重要な課題である。

内田 G と小畠 G は、自らが発見した上記の現象に基づき、新たなフォトメカニカル現象の開拓を目的に研究を展開した。小畠 G はマクロスケール（数 mm 以上）のジアリールエテン誘導体単結晶の光誘起結晶形状変化を対象として、定常光照射によって通常の光誘起屈曲の後、屈曲が加速する特異的二段階光屈曲挙動を見いだすとともに、動的挙動と結晶格子の詳細な解析を行い、分子反応から巨視的応答を導く階層的機構を解明した（*Nanoscale* 2018）。これらの知見に基づき、光の照射方向に依存して屈曲、円筒状螺旋およびねじれ挙動を示す結晶の開発に成功し、結晶形態の精密運動光制御を可能とした（図 15, *JACS* 2018, Front Cover）。

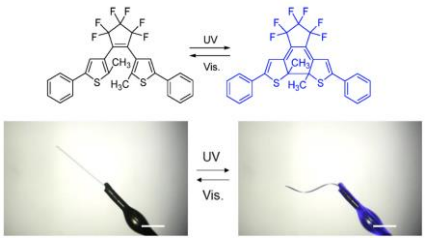

図 15. 結晶形態の精密運動光制御.

内田 G では、光照射による分子異性化反応と相分離挙動を利用した結晶成長により、ハスの葉のダブルラフネス構造を模した表面を作成し、可逆的な光制御超撥水性表面を創製した（*JACS* 2016）。さらに、フォトサリエント（光粉砕）現象を示す中空結晶を見いだし、直径 1 ミクロンのビーズを内包させた結晶に紫外光照射することで、その内包物の放出が可能なメゾスコピック系を構築した（図 16、*ACIE* 2017, Hot Paper, 森本 G との共同研究）。またマクロスコピック系の高度光運動展開にも取り組み、繊毛運動を模倣した数 mm サイズのマクロスコピック光輸送可能なスマート表面を構築した（図 17、*ACIE* 2019, Front Cover, 宮坂 G との共同研究）。

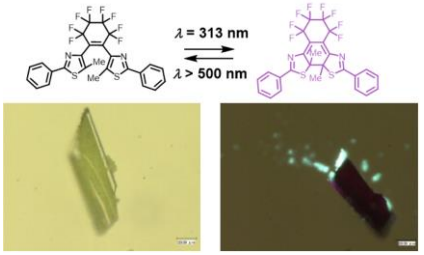

図16. フォトサリエント現象.

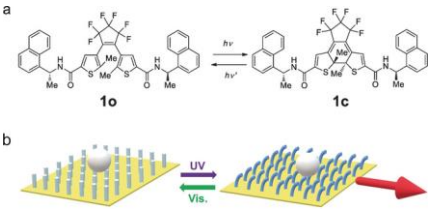

図17. 繊毛運動を模倣したマクロスコピック光輸送スマート表面.

則包 G ではアゾベンゼンモノマーを含む共重合高分子を対象に、光異性化反応の進行に伴うガラス転移温度の上下を利用し、微弱光で変形駆動する高感度光メカニカル応答を実現した（図 18, *Nature Comm.* 2018）。

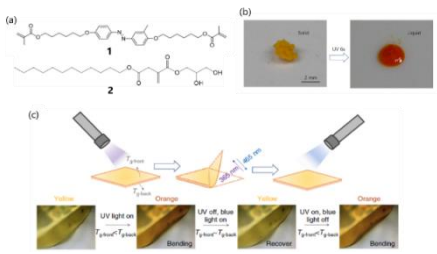
図 18. 光反応によるマクロ物質輸送計の構築. 達成：則包.

また景山 G はアゾベンゼン混合共結晶を用い、マクロスコピックサイズの集合体において、連続光照射下で周期的

な自発励振系の作製に成功した（図 19, *ACIE*, 2016）。

池田 G はアゾトラン誘導体を有する架橋液晶高分子を用い、２光子吸収による多様な形態変化の精密制御を実現した（*J. Mat. Chem. C* 2015, *Mol. Cryst. Liq. Cryst.* 2018, 宮坂 G との共同研究）。

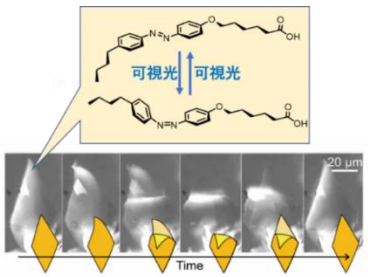

図 19. 光照射により自律的振動運動を行うマクロスコピック集合体.

森本 G と阿部 G ではフォトクロミック分子を内包するサブミクロンからミクロンサイズの高分子ビーズを対象に、宮坂 G と共同してレーザートラッピング下においてフォトクロミック反応に伴う μm から 10μm 程度のビーズの可逆的並進移動を実証し、分子レベルの反応をメゾスコピック系の応答に展開する新手法を開発した（図 20, *JPCL* 2018, *JPCC* 2018 *Front cover, Editor's Choice*）。

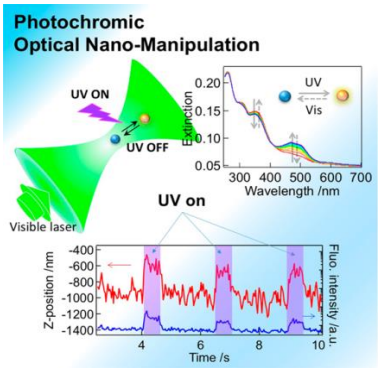

図 20. 光反応を用いたレーザートラッピング下の μm サイズ粒子の可逆的並進運動.

<u>固体系光ダイナミクスの計測法開発と応用</u>

種々のサイズの固体や集合体系における分子間の協同効果が顕著に発現する複合光応答の機構解明には、新たな時間・空間計測手法の開発と応用も重要な役割を果たす。

朝日 G では、フェムト秒顕微ダイナミクス測定装置を開発し（*JPCL* 2016）、水中に分散した粒径 50 nm の C_{60} ナノ結晶を対象として、フェムト秒パルスレーザーで励起により隣接した分子間の電荷移動（CT）を経た励起状態（C_{60}^{+}-C_{60}^{-}）の生成を見出し、強励起条件では隣接する CT 励起状態間で、励起子消滅が抑制されていることを明らかにし（図 21、*PCCP* 2018、宮坂 G との共同研究）、集合体における多励起子利用につながる結果を示した。また Vacha G では、エネルギー供与体と三重項増感剤を少量添加したエネルギー受容体の多結晶を用いて、三重項消滅（TTA）に基づくアップコンバージョン蛍光を点光源として利用することで、多結晶中の三重項励起子の拡散過程を光の回折限界よりも小さな領域で可視化することに成功し（*Nanoscale* 2017）、集合体系の励起子移動に関わる高空間分解計測を可能とした。

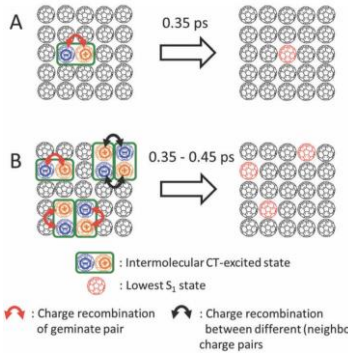

図 21. C_{60} ナノ結晶の光応答. (A) 弱励起, (B) 強励起条件

以上のように複合光応答系の機能開拓と機構解明による学理の解明、新規複合応答を可能とする光強度に閾値を持つ分子系の開発、測定手法の開発など、当初の目的に沿った結果を得た。

その他にも、本新学術領域の期間中に数多くの化合物や計測手法も開発された。

鳥本 G では、化学組成、構造の精密制御された QD（半導体量子ドット）の作製を行い、多励起子利用への展開を可能とした（ *J. Phys. Chem. C* 2015, *ACS Appl. Mater. Interfaces* 2018）。また、化合物のみならず、集合体としても多様な物質群が開発されたことは、記述の通りである。

計測手法に関連したトピックスとしては、森本 G で開発された超解像蛍光顕微鏡用の蛍光スイッ

チング（開環体では蛍光 OFF、閉環体で ON 状態となる）を示すジアリールエテン誘導体があげられる。この誘導体では、開環体の 0-0 バンドよりも長波長域に存在するホットバンド（Urbach tail）を可視光励起することにより閉環反応が誘起され、蛍光性閉環体の生成が可能であることが明らかになった（*JACS* 2017, 宮坂 G との共同研究）。この蛍光性閉環体は同じ可視光照射により無蛍光性の開環体に戻ることから、光異性化反応に伴う蛍光の ON-OFF、OFF-ON、および蛍光励起の3過程を可視単一波長レーザー光で担う超解像蛍光イメージングが可能となり（*Chem. Com.* 2017, *Back Cover,* 宮坂 G との共同研究）。また、伊藤 G は固体集合過程の階層的ダイナミクスの蛍光イメージング測定手法の開発（*Sci. Rep.* 2016, *PPS* , 2016 前田 G との共同研究）なども行われた。

まとめと展望

本新学術領域では、既に述べたように ① 超高速な高位電子励起状態からの緩和、② 迅速な励起子消滅、③ 限定された光学遷移可能電子状態 の 3 種の制限を超え、多数光子や多数励起分子による分子・分子集合体の高次複合光応答の開拓を目的とした。A01 班ではこれらの 3 種の制限に対して、正面突破を目指した研究を、A02 では、励起子消滅による“量の損失”を超えるための多分子協調による加算・増幅的複合応答系の開拓を、A03 では、多数光子や多数励起分子によるメゾ、マクロ複合応答へとつながる光応答系の開拓、機構解明、高度展開を行った。

上に示した① と ② の制限の超克のためには、これらの高速過程を超える超高速反応を誘起することこそが、最も単純な方法である。しかし、サブピコ秒（～10^{-13} 秒）程度の高位電子励起状態の寿命は、分子の大きな構造変化に相当する低波数分子内振動の 1 周期以下、また溶媒など媒体の揺らぎと同程度以下の時間スケールである。そのため、（擬似）平衡状態を基本とし、分子内、分子間の揺らぎと活性化エネルギー等を用いた既存の化学反応理論からは、このような高速反応を予測することは困難となる。したがって、原理は明確であっても、現実的には研究の方法論の開発が重要な課題となる。

既述のように①に関しては、電子状態や核配置を選択した場合には、10^{-13} 秒程度の短い時間であっても異性化や電子移動反応を効率的に誘起可能であることが、実験的に示されるとともに、高位電子励起状態の計算手法も開発され、今後は、理論と実験両面から効率的に高位電子励起状態からの種々の反応や応答を探索可能となることが期待できる。

次に、順番は前後するが、③について述べる。③ の多様性の制限に関わる課題としては、多光子吸収や多重励起により顕在化する禁制電子状態からの反応があげられる。これについては、①にも関連し、特に軌道対称性と反応性の観点からも合理的な解釈が可能な例が提出された。一方、プラズモン等を利用した強結合場では、摂動を超えた新規電子状態の創出が可能であり、その選択則を含めて基礎的な知見が明らかになると共に、強結合場の応用による高効率光反応・応答系の作成もなされた。これらの原理を基に、新たな展開が可能となると考えている。

励起子融合や分裂の過程は、② に直接関わる課題の一つである。本新学術領域において、その詳細な機構が理論的に解明されると共に、これに基づく高効率系が A01、A02 からも実現され、

今後は、いかに融合あるいは分裂した励起子間の消滅を防ぐかを含め、理論・実験による研究の発展が望まれる。

また　②　の超克に対しては、“高密度励起を避ければ励起子消滅は進行しない” という解法も存在する。実験的には A02 や A03 班を中心とした多くの研究から、通常の光源照射（弱励起条件）による多数の光子や多数の分子の光反応によって、集合体全体やマクロサイズの構造変形などの光応答が観測された。これら弱励起条件で起こる分子個々の光反応によって集合体やメゾマクロスコピックのスケールの応答が進行する場合には、個々の分子の反応の結果が系に “蓄積” されるとともに、その蓄積により集合体の熱力学的安定性の変化が誘起されることが重要な因子として作用している場合が多い。この安定性の変化により、集合体の相転移や相分離などが誘起され、メゾ、マクロサイズの光応答が進行する。また、相転移や相分離が進行しない、あるいはできない場合でも、結晶格子のひずみなどによるマクロサイズの光応答が観測される系も存在する。特に構成する分子が２つ以上の状態間で可逆的な光反応を行う場合には、集合体やマクロサイズの物質の光応答が可逆的に進行する場合も多い。集合体全体の安定性が、構成する可逆光反応分子の２状態の組成に大きく依存すれば、集合体の可逆的変化を誘起できることは原理的には自明であり、今後の高次複合光応答を発展のための重要な原理を与えている。

また連鎖反応系のネットワーク作成が重要な課題となる増幅光応答系においても、特に光で容易に開始できる系として分子内異性化反応系を用いると、異性体 A と B の間でいくつかの物性の安定性が異性体間で逆転関係を持つ場合には、合理的な連鎖反応系の設計・構築が可能であることも示されており、今後の展開に対する原理的指針となっている。

以上のように、個々の計画班・公募班のみならず領域内や国際共同研究、また領域会議やシンポジウムでのディスカッションなどにより、高次複合光応答に関わる新現象の開拓、機構や原理の解明による学理の構築、これらを利用した新規光反応系への展開がなされるとともに、今後の研究の方法論も提示できたと考えられる。

これらの内容についての詳細は、“Photosynergetic Responses in Molecules and Molecular Aggregates” として Springer 社から出版（2020 年 9 月予定）される。