### **Exploring Quantum-Classical Boundary**

### **Department of Photo-Molecular Science Division of Photo-Molecular Science II**



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#### Education

	B. E. The University of Tokyo	
1992	Ph.D. The University of Tokyo	
Professional Employment		
1992	Research Associate, Tohoku University	
2001	Associate Professor, Tohoku University	
2003	Professor, Institute for Molecular Science	
	Professor, The Graduate University for Advanced Studies	
2004	Visiting Professor, Tohoku University ( –2005)	
2007	Visiting Professor, Tokyo Institute of Technology ( –2008)	
2009	Visiting Professor, The University of Tokyo (-2011)	
2012	Visiting Professor, University of Heidelberg	
2014	Visiting Professor, University of Strasbourg ( –2016)	
Awards		
1998	Award by Research Foundation for Opto-Science and	
	Technology	
2007	JSPS Prize	
2007	Japan Academy Medal	
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- 2009 Fellow of the American Physical Society
- 2012 Humboldt Research Award
- 2017 Hiroshi Takuma Memorial Prize of Matsuo Foundation
- 2018 Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology of Japan

Keywords

Quantum-Classical Boundary, Quantum Technology, Coherent Control

It is observed in a double-slit experiment by Tonomura and coworkers that single electrons recorded as dots on a detector screen build up to show an interference pattern, which is delocalized over the screen.<sup>1)</sup> This observation indicates that a delocalized wave function of an isolated electron interacts with the screen, which is a bulk solid composed of many nuclei and electrons interacting with each other, and becomes localized in space. This change, referred to as "collapse" in quantum mechanics, is often accepted as a discontinuous event, but a basic question arises: When and how the delocalized wave function becomes localized? Our dream is uncovering this mystery by observing the spatiotemporal evolution of a wave function delocalized over many particles interacting with each other. Having this dream in mind, we have developed coherent control with precisions on the picometer spatial and attosecond temporal scales. Now we apply this ultrafast and ultrahigh-precision coherent control to delocalized wave functions of macroscopic many-particle systems such as an ensemble of ultracold Rydberg atoms, as depicted schematically in Figure 1, and a bulk solid, envisaging the quantum-

#### Selected Publications

- H. Katsuki et al., "Visualizing Picometric Quantum Ripples of Ultrafast Wave-Packet Interference," Science 311, 1589-1592 (2006)
- H. Katsuki et al., "Actively Tailored Spatiotemporal Images of Quantum Interference on the Picometer and Femtosecond Scales," Phys. Rev. Lett. 102, 103602 (2009).
- K. Hosaka et al., "Ultrafast Fourier Transform with a Femtosecond-Laser-Driven Molecule," Phys. Rev. Lett. 104, 180501 (2010).

classical boundary connected smoothly.



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Figure 1. Schematic of the many-body system of ultracold Rydberg atoms 2

- H. Goto et al., "Strong-Laser-Induced Quantum Interference," Nat. Phys. 7, 383-385 (2011).
- H. Katsuki et al., "All-Optical Control and Visualization of Ultrafast Two-Dimensional Atomic Motions in a Single Crystal of Bismuth," Nat. Commun. 4, 2801 (2013).
- N. Takei et al., "Direct Observation of Ultrafast Many-Body Electron Dynamics in an Ultracold Rydberg Gas," Nat. Commun. 7, 13449 (2016).

# 1. Ultrafast Coherent Control of Condensed Matter with Attosecond Precision<sup>3)</sup>

Coherent control is a technique to manipulate wave functions of matter with light. Coherent control of isolated atoms and molecules in the gas phase is well-understood and developed since the 1990s, whereas its application to condensed matter is more difficult because its coherence lifetime is shorter. We have recently applied this technique to condensed matter samples, one of which is solid para-hydrogen (p-H<sub>2</sub>). Intramolecular vibrational excitation of solid p-H<sub>2</sub> gives an excited vibrational wave function called a "vibron," which is delocalized over many hydrogen molecules in a manner similar to a Frenkel exciton. It has a long coherence lifetime, so we have chosen solid p-H<sub>2</sub> as our first target in the condensed phase. We shine a time-delayed pair of femtosecond laser pulses on p-H<sub>2</sub> to generate vibrons. Their interference results in modulation of the amplitude of their superposition. Scanning the interpulse delay on the attosecond time scale gives a high interferometric contrast, which demonstrates the possibility of using solid p-H2 as a carrier of information encoded in the vibrons.

In the second example, we have controlled the terahertz collective phonon motion, called a "coherent phonon," of a single crystal of bismuth. We employ an intensity-modulated laser pulse, whose temporal envelope is modulated with terahertz frequency by overlap of two positively chirped laser pulses with their adjustable time delay. This modulated laser pulse is shined on the bismuth crystal to excite its two orthogonal phonon modes. Their relative amplitudes are controlled by tuning the delay between the two chirped pulses on the attosecond time scale. Two-dimensional atomic motion in the crystal is thus controlled arbitrarily. The method is based on the simple, robust, and universal concept that in any physical system, two-dimensional particle motion is decomposed into two orthogonal one-dimensional motions, and thus, it is applicable to a variety of condensed matter systems.



**Figure 2.** Schematic diagram of the experimental setup for the observation of ultrafast many-body electron dynamics in a strongly correlated ultracold Rydberg gas.<sup>3)</sup>

In the third example, the double-pulse interferometry used for solid p-H<sub>2</sub> has been applied to many-body electronic wave functions of an ensemble of ultracold rubidium Rydberg atoms, hereafter called a "strongly correlated ultracold Rydberg gas," as shown in Figure 2. This has allowed the observation and control of many-body electron dynamics of more than 40 Rydberg atoms interacting with each other. This new combination of ultrafast coherent control and ultracold atoms offers a versatile platform to precisely observe and manipulate nonequilibrium dynamics of quantum many-body systems on the ultrashort time scale.

# 2. Attosecond Control of Electronic Structure Symmetry Restoration<sup>4)</sup>

Laser pulses can break the electronic structure symmetry of atoms and molecules by preparing a superposition of states with different irreducible representations. Here, we discover the reverse process, symmetry restoration, by means of two circularly polarized laser pulses. The laser pulse for symmetry restoration is designed as copy of the pulse for symmetry breaking. Symmetry restoration is achieved if the time delay is chosen such that the superposed states have the same phases at the temporal center. This condition must be satisfied with precision of a few attoseconds. Numerical simulations are presented for the C<sub>6</sub>H<sub>6</sub> molecule and <sup>87</sup>Rb atom as shown in Figure 3. The experimental feasibility of symmetry restoration is demonstrated by means of high-contrast time-dependent Ramsey interferometry of the ultracold <sup>87</sup>Rb atom.



**Figure 3.** Numerical simulations of the evolutions of electronic densities upon laser-induced symmetry breaking, charge migration, and symmetry restoration in the (a)  $C_6H_6$  molecule and (b) <sup>87</sup>Rb atom. The central panels show snapshots of the time-dependent parts of their electronic densities.<sup>4</sup>)

#### References

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K. Ohmori, *Found. Phys.* **44**, 813–818 (2014).
H. Katsuki *et al., Acc. Chem. Res.* **51**, 1174–1184 (2018).
C. Liu *et al., Phys. Rev. Lett.* **121**, 173201 (2018).

### Awards

OHMORI, Kenji; Hiroshi Takuma Memorial Prize of Matsuo Foundation (2017). OHMORI, Kenji; Commendation for Science and Technology by the Minister of Education, Culture, Sports, Science and Technology of Japan (2018).

- \* IMS International Internship Program
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