Review

Development of ultrahigh-precision coherent control and its applications

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(Communicated by Toshimitsu YAMAZAKI, M.J.A.)

Abstract: Coherent control is based on optical manipulation of the amplitudes and phases of wave functions. It is expected to be a key technique to develop novel quantum technologies such as bond-selective chemistry and quantum computing, and to better understand the quantum worldview founded on wave-particle duality. We have developed high-precision coherent control by imprinting optical amplitudes and phases of ultrashort laser pulses on the quantum amplitudes and phases of molecular wave functions. The history and perspective of coherent control and our recent achievements are described.

Keywords: wave packet, coherent control, quantum interference, laser, attosecond, femtosecond

1. Introduction

The wave nature of matter is at the heart of the quantum world. It is almost 80 years since quantum mechanics was established in the early 20$^{\text{th}}$ century, and our modern societies are deeply indebted to inventions made possible by quantum mechanics. The worldview of quantum theory based on the wave-particle duality is, however, still hard to be smoothly connected to our ordinary one, and the wave nature of matter is not fully utilized in modern technology. We are trying to control completely the wave nature of matter to better understand quantum theory. Improved understanding of quantum theory will result in the development of novel quantum technologies such as bond-selective chemistry and quantum computing. Such control is called "coherent control." Coherent control is based on optical manipulation of quantum amplitudes and phases of wave functions. It is a basic scheme of controlling a variety of quantum systems from simple atoms to nanostructures. One promising approach is to use coherent light to modulate a matter wave with its optical phase. Let us take an example where a diatomic molecule is irradiated with an ultrashort optical pulse resonant with its vibronic transitions, as schematically shown in Fig. 1. If the temporal width of the pulse is on the order of femtoseconds (fs) and is shorter than the classical vibrational period of the molecule, which is typically on the order of fs to picoseconds (ps), the bandwidth of the pulse is then broad enough to be resonant simultaneously with the transitions to plural vibrational eigenstates. Those eigenstates are consequently superposed and interfere with each other to generate a spatially localized atom wave called a "wave packet (WP)." Since the temporal oscillation periods of the eigenstates within the WP are different from one another, the WP moves back and forth along the bond axis, corresponding to the classical vibrational motion of the molecule. In the generation of this WP, the optical amplitude and phase of a particular frequency component of the pulse is imprinted on the corresponding eigenstate as its quantum amplitude and phase. Hence manipulating the optical amplitude and phase allows us to manipulate the quantum amplitude and phase of the WP. This is a schematic description of the concept of coherent control. It is seen here that this

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doi: 10.2183/pjab/84.167

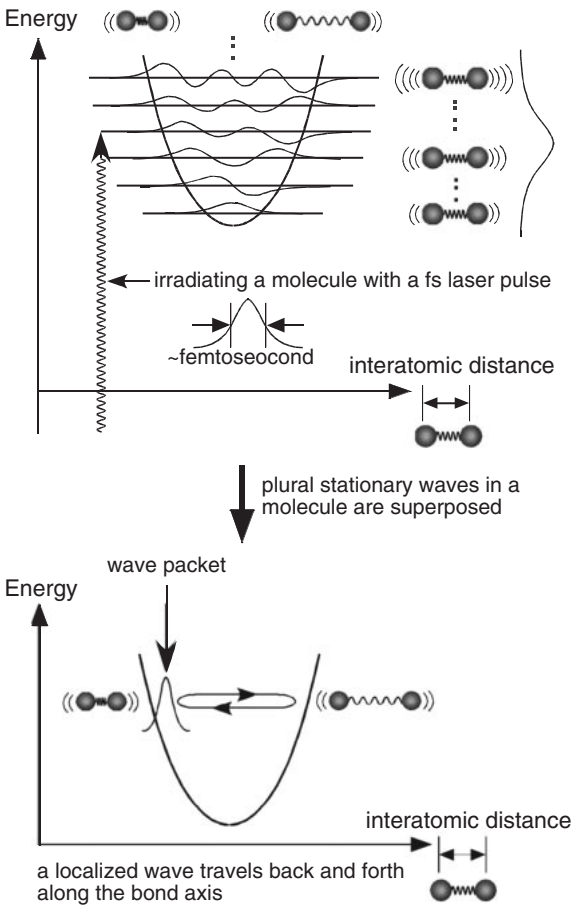

Fig. 1. Schematic representation of the generation of a vibrational wave-packet with a femtosecond optical pulse.

concept is basically applicable not only to diatomic molecules but also to any quantum system with any type of eigenstates. In this review, I describe the history of coherent control and our recent achievements in this research field.

2. History of coherent control

The concept of coherent control was first proposed in the middle of 1980's by theoretical chemical physicists to control chemical reactions. Paul Brumer and Moshe Shapiro published seminal papers where they described the scenario to have constructive and destructive interferences in the desired and undesired reaction pathways, respectively.[1),2)] For example, when a molecule is irradiated with a laser beam and its third harmonic simultaneously, quantum interference occurs among vibronic wave-functions whose oscillations are synchronized with those of the two laser fields. It may be, therefore, possible to induce particular interference to promote a specific dissociation pathway by tuning the relative phase of those two laser fields. This kind of scenario, where a molecule is irradiated simultaneously with a laser beam and its harmonic, is categorized as Brumer-Shapiro type, and later has been realized experimentally for the ionization of the Hg atom[3)] and for the dissociation and ionization of the HI molecule,[4),5)] for example. On the other hand, David J. Tannor, Ronnie Kosloff, and Stuart A. Rice proposed an alternative scenario where a couple of fs laser pulses are employed.[6),7)] In this scheme, a molecule is irradiated with a fs laser pulse to create a WP on the excited potential surface, where the WP travels to reach a particular point to be further excited or de-excited by another fs laser pulse to a dissociative surface. By tuning the timing of those two fs laser pulses on the time scales of nuclear motion, say, tens of femtoseconds to picoseconds, a specific dissociation pathway could be selected. This kind of scenario is called Tannor-Rice type, and later has been experimentally demonstrated for Na_2[8)] and Xe-I_2,[9)] for example. In early 1990's, Graham R. Fleming and his coworkers demonstrated another approach where the interference of two molecular WPs was controlled with a pair of fs laser pulses.[10),11)] This approach is now called "wave-packet interferometry (WPI)," and our recent experimental achievements are also based on this WPI scheme. Its history and its cutting edge that we have recently demonstrated will be described in the following sections. For any of these control schemes mentioned above, their experimental implementations were not made until 1990's when the laser technologies was rapidly developed. Recently, modern optical technologies such as ultrashort-pulse lasers and pulse shapers have accelerated the experimental development of coherent control. Moreover, the closed-loop control of pulse shapes based on the genetic algorithm has been found to be effective in controlling the dynamics of large complex molecules.[12)–16)] The concept of coherent control is cross-disciplinary now, and the range of its applications includes a variety of fields such as solid-state dynamics[17)–21)] and information processing with quantum states of matter.

3. Wave-packet interferometry

A pair of electronic or vibrational WPs can be

created in a single atom or molecule with a pair of femtoseocnd laser pulses, and interfere with each other. By controlling the relative phase of the fs pulses, constructive or destructive interference can be selectively induced to those two WPs. This is a schematic concept of WPI. WPI is one of the most fundamental and important scenarios of coherent control where the essentials of Brumer-Shapiro type and Tannor-Rice type have been combined. WPI was first demonstrated by Graham R. Fleming and his co-workers in the early 1990's.[10),11)] They employed a pair of fs laser pulses whose relative phase was locked to create a pair of phase-locked vibrational WPs in the iodine molecule (I_2). WPI has so far been developed with Rydberg and spin-orbit WPs in atoms and molecules[22)–41)] and vibrational and rotational WPs in molecules[10),11),42)–52)] based on the measurement of population of the bound excited state sensitive to the relative phase of the twin optical pulses. Aside from such bound-state interferometry, WP interference has also been observed for the free states located above the dissociation limits of the relevant potential curves.[53)–56)] Similar double pulse approaches have also been applied to condensed phases.[17)–21)] Recently, novel efforts have been made with WPI to read and write information by controlling the amplitudes and phases of Rydberg states of an atom. Such information processing should benefit from a molecule with more degrees of freedom of internal quantum states than an atom, say, vibration and rotation. This type of information processing is, on the other hand, carried out with an ensemble of atoms or molecules, and more degrees of freedom in a molecule can be a source of serious quantum dephasing of a molecular ensemble. The thermal distribution of rotational states is, for example, known to be a major source of dephasing in molecular systems that can wipe out the interference structure much more quickly than in atomic systems.[10),11),42)–44)] We succeeded in drastically reducing this rotational dephasing by employing a cold and dilute ensemble of the molecules prepared by the supersonic jet expansion and narrow-band interrogation of the state populations with a nanosecond (ns) probe laser pulse.[46)] We have also succeeded in locking the relative phase of two fs laser pulses (typically $\sim$ 100–200 fs pulse-width) by using our homemade highly-stabilized optical interferometer called "attosecond phase modulator (APM)."[49),51)] These two advances have been combined to realize unprecedentedly high-precision WPI, which has been further utilized to read and write quantum codes composed of the amplitudes and phases of molecular vibrational eigenstates.[49),51)]

4. Controlling quantum ripples with light

Figure 2(a) schematically shows interference of two counterpropagating vibational WPs in the I_2 molecule. It is seen that picometric ripples appear only when the two WPs cross each other. The lifetime of each ripple is no more than $\sim$ 100 fs. We have recently succeeded in visualizing these quantum ripples, as shown in the left panel of Fig. 2(b).[57)] In this visualization we employed a pump-probe scheme shown in Fig. 3 where we used a probe pulse to excite the quantum ripple generated on the B-state potential curve to the upper E-state one. The fluorescence intensity from this E-state was measured as a function of the timing τ_{probe} and the wavelength λ_{pr} of the probe pulse. Based on the classical Franck-Condon principle, it is reasonable to consider that the E-B excitation occurs around a particular internuclear distance where the energy of the probe pulse is equal to the difference of the potential energies of the E and B states, which is a function of the internuclear distance r. The wavelength λ_{pr} is, therefore, successfully converted to the internuclear-distance r. We have thus obtained the space-time image shown in the left panel of Fig. 2(b), where we measured eight quantum beats by scanning τ_{probe} with eight different λ_{pr}'s, and they were interpolated along the λ_{pr} axis.

Theory predicts that the space-time image shown in Fig. 2(b) can be actively designed if we can precisely tune the relative phase of the two counterpropagating WPs schematically shown in Fig. 2(a). Figure 4 shows the pump-control-probe scheme to demonstrate such actively designed space-time images. We have utilized APM to generate a phase-locked fs pump and control laser pulses (typically $\sim$ 100–200 fs pulse-width) to be used to generate a pair of phase-locked WPs on the B-state potential curve. APM is a Michelson interferometer assembled in a vacuum chamber to reduce the influence of air-flow disturbance and temperature drift. A mechanical stage is installed in one arm of APM for coarse setting of the delay τ_{control} between the pump and control pulses, and

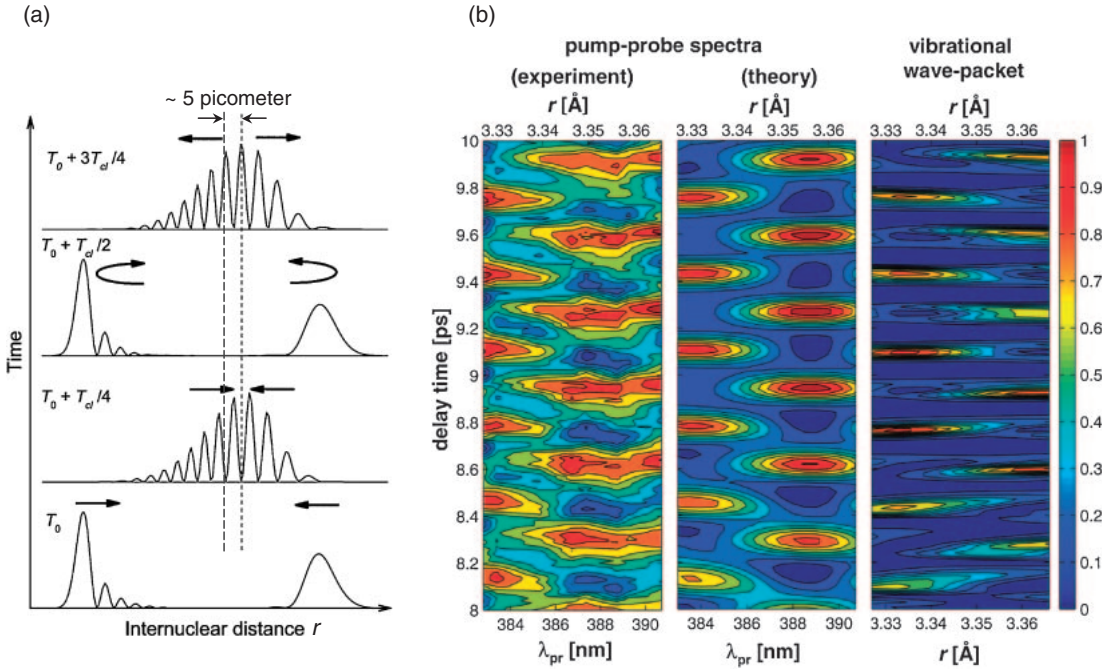

Fig. 2. Quantum ripple. (a) Schematic representation of the interference of two vibrational wave-packets counterpropagating in the I_2 molecule. A picometric quantum ripple transiently appears only when the two wave packets cross each other. T_{cl} is a classical vibrational period of the molecule and is around 0.3 ps. (b) Quantum ripples in the I_2 molecule visualized experimentally (left), the theoretical simulation of the experimental signal (middle), and the theoretical simulation of the wave packet (right). Adopted from Science **311**, 1589–1592 (2006). λ_{pr} and r denote the wavelength of the probe pulse and the internuclear distance, respectively.

the other arm has a gas cell for the pressure tuning of $\tau_{control}$ with the attosecond stability and resolution. Figure 5 shows an optical interferogram of the pump and control pulses tuned to ~ 537 nm. In this measurement, $\tau_{control}$ was tuned around ~ 0 fs so that the pump and control pulses were temporally superimposed. The total intensity of these superimposed pulses was measured while the argon pressure within the gas cell was scanned, and was plotted against $\tau_{control}$ which was calibrated by an optical cycle of the present carrier wavelength ~ 537 nm. The origin $\tau_{control} = 0$ is arbitrary. One standard deviation of the sine curve least-square-fitted to the measured optical interferogram is only 700 zeptoseconds. During the measurement of the quantum beat as a function of the delay τ_{probe} of the fs probe pulse, $\tau_{control}$ was actively stabilized on the attosecond scale with periodical compensations of the drift of a spectral interferogram of the pump and control pulses monitored with a linear detector attached to a spectrometer. Figure 6 shows a snapshot that I have taken in my laboratory. The image projected on the front wall is the spectral interferogram of the pump and control pulses. The horizontal drift of this interferometric spectrum from peak to peak corresponds to the π drift of the pump-control phase, which corresponds to ~ 1.8 fs in $\tau_{control}$ in the present case. It is seen in this picture that the spectrum is not blurred even though the images of the active members of my group are completely blurred. By using this APM, we have succeeded in actively designing the space-time image of the WP interference very recently.[58)]

5. Information processing with light and molecules

Ultrahigh-precision WPI described above has allowed to encode information into the amplitudes and phases of vibrational eigenstates within WPs. The experimental scheme is again shown in Fig. 4, where we use both of the fs and ns probe pulses this time. Information is encoded by the interference of two WPs generated with the pump and control

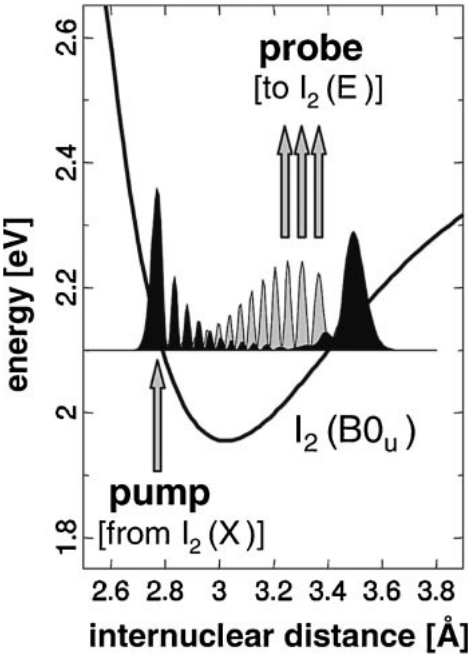

Fig. 3. Pump-probe scheme to visualize quantum ripples in the I_2 molecule. A quantum ripple generated on the B-state potential curve is further excited by the probe pulse to the upper E state, and the fluorescence from the E state is monitored. A particular part along the internuclear distance is selectively probed by tuning the wavelength of the probe pulse, based on the classical Franck-Condon principle successfully applied to the E-B excitation. Adopted from Science **311**, 1589–1592 (2006).

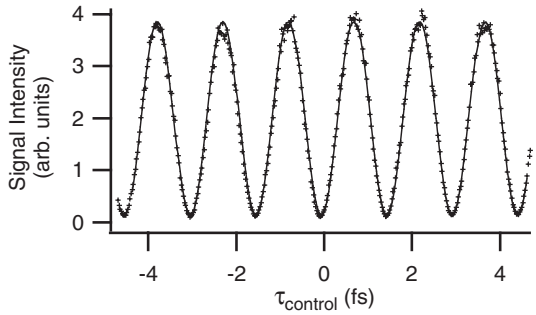

Fig. 5. Example of the optical interferogram of the pump and control pulses tuned around 537 nm measured with the attosecond phase modulator (APM) by scanning $\tau_{control}$ around 0 fs. The origin $\tau_{control} = 0$ is arbitrary. The abscissa was calibrated by an optical cycle of the present carrier wavelength. One cycle of the oscillation corresponds to 1.79 fs. The solid curve represents a sine curve least-square-fitted to the measured interferogram. One standard deviation of the oscillation period is 700 zeptoseconds. Reprinted figure with permission from H. Katsuki *et al.*, Phys. Rev. A **76**, 013403 (2007). Copyright (2007) by the American Physical Society.

pulses and decoded with the fs and ns probe pulses. Figure 7(a) shows examples of the codes retrieved as the squares of the amplitudes of the vibratinal eigenstates measured by scanning the wavelength of a narrow-band ns pulsed laser. We call these codes "population codes." Population codes show only amplitude information. Phase information is also

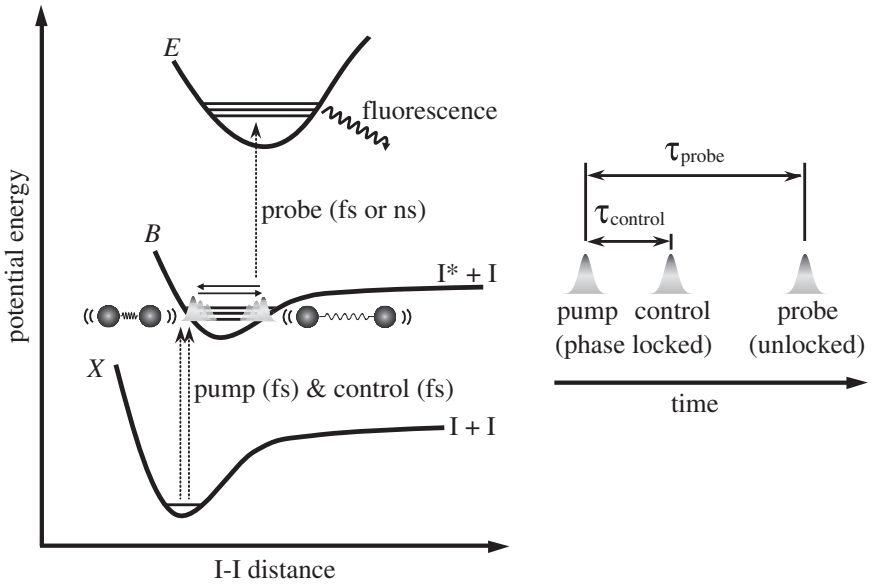

Fig. 4. Pump-control-probe scheme for the real-time or state-resolved measurements of wave-packet interference with the fs or ns probe pulse. The potentials are only schematic. Reprinted figure with permission from K. Ohmori *et al.*, Phys. Rev. Lett. **96**, 093002 (2006). Copyright (2006) by the American Physical Society.

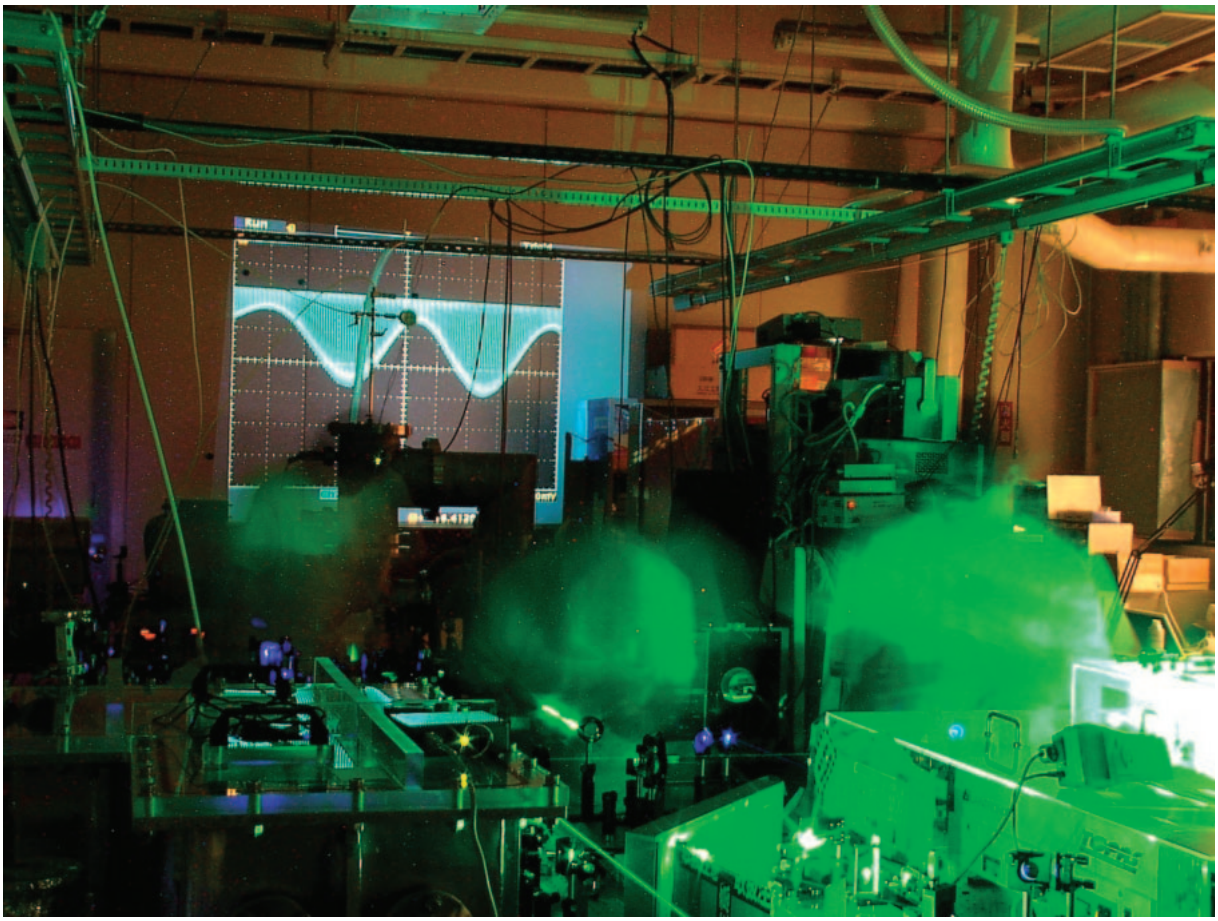

Fig. 6. Snapshot in my laboratory. The vacuum chamber seen in the lower left hand corner is the attosecond phase modulator (APM). The image projected on the front wall is the interferometric spectrum of two fs laser pulses generated by APM. The horizontal drift of this spectrum from peak to peak corresponds to ~ 1.8 fs drift in the interpulse delay.

necessary to retrieve all the quantum information stored in WPs. Phase information is the information on the relative timing of the temporal oscillations of the vibrational eigenstates involved in the WPs, and it should, therefore, appear in the temporal evolutions of WPs. Figure 7(b) shows quantum beats representing such temporal evolutions measured by scanning the delay τ_{probe} of the fs probe pulse. We call these temporal evolutions "phase codes." Although population codes A and C are barely distinguishable, phase codes A and C are in anti-phase to be clearly distinguished from each other. This π phase shift of phase codes arises from the π phase shift of the even vibrational eigenstates relative to the odd ones. We have experimentally demonstrated that a combination of amplitude and phase codes has allowed us to read and write quantum information with molecular vibrational WPs.[49),51)] The next step should be the development of quantum gates. We have proposed Quantum Fourier Tranfrorm (QFT) based on the temporal evolution of the vibratinal WPs in the I_2 molecule and have tested its experimental feasibility with theoretical simulations.[59)] Very recently we have succeeded in experimental implementation of this WP-based QFT.[60)] QFT is the key to Shor's algorithm for factoring large integers,[61)] and intensive efforts have been made recently on its experimental realization.[62),63)]

6. Future perspective

Scalability is a key issue to be considered for further promoting quantum information processing with molecular WPs.[49),51),64)–66)] Theoretical studies

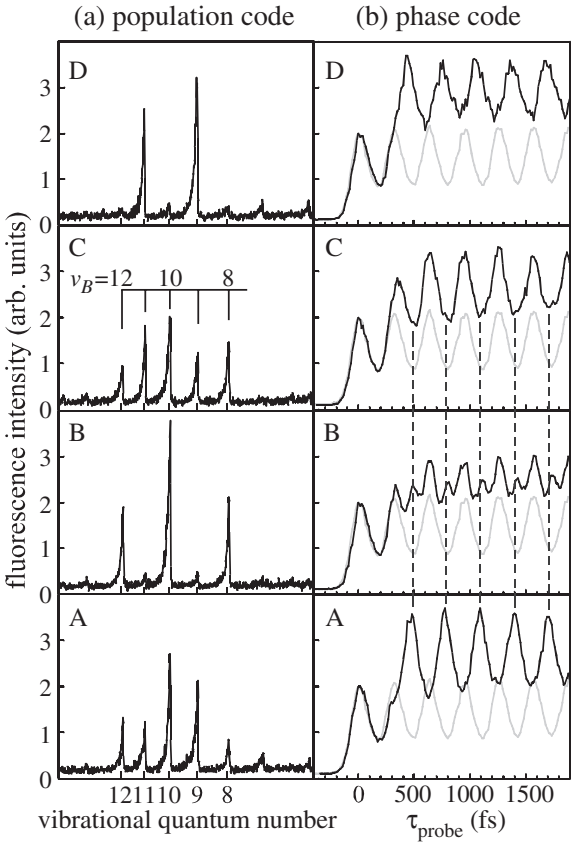

Fig. 7. Quantum code encoded in the vibrational wave-packets of the iodine molecule with a pair of fs laser pulses (pump and control pulses) produced by APM. (a) Populaiton code. The population distribution of the vibrational eigenstates within the wave packets is measured by scanning the wavelength of the ns probe pulse. The relative phase of the pump and control pulses is increased in steps of $\sim \pi/2$ in going from code A to code D. (b) Phase code. Real-time evolutions of the wave-packet interference measured with the same pump-control phases as for population codes A-D. Reprinted figure with permission from K. Ohmori *et al.*, Phys. Rev. Lett. **96**, 093002 (2006). Copyright (2006) by the American Physical Society.

are in progress to address this challenging subject.[67)]

In the microscopic world of atoms and molecules, the interference of quantum waves is naturally observed. The target of coherent control is no longer restricted to isolated atoms and molecules in the gas phase, but now being extended to more complex systems such as the liquid and solid phases. Recently the role of coherence has begun to be investigated in biological systems such as visions[16)] and photosynthesis.[68)] The biological system may be the future target of coherent control. These complex systems always suffer from interactions among many atoms and molecules, so that the wave nature of matter is lost more quickly than the isolated system. This is called "decoherence." The test and management of decoherence should be challenged in the next step. We have already started experimental efforts in this direction. On the other hand, quantum theory is even now a shocking theory. Not a few people are still uncomfortable with wave-particle duality. As long as coherent control utilizes wave nature of matter, pursuing coherent control must be an occasion to test the quantum worldview. On the way to our goal, we may find a hint to better understand the quantum world.

Acknowledgement

The author acknowledges all the collaborators who coauthored the papers introduced in this review. Especially H. Katsuki and H. Chiba (Institute for Molecular Science) have played indispensable roles in starting and developing this challenging and difficult research. This work was partly supported by Grant-in Aid from MEXT of Japan.

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(Received Feb. 19, 2008; accepted Mar. 31, 2008)

Profile

Kenji Ohmori was born in 1962 in Kumamoto, Japan. After he received his Doctor of Engineering from the University of Tokyo in 1992 under the direction of Professor Hiroyuki Matsui, he started his research career as Research Associate with Professor Yukinori Sato at Tohoku University, and was promoted to Associate Professor in 2001. In 2003 he moved to Institute for Molecular Science to start his own research group as Full Professor, and was appointed to Director of Laser Research Center for Molecular Science in 2007. He was Visiting Professor at Tohoku University (2004–2005) and Tokyo Institute of Technology (2007–2008). He was awarded by Research Foundation for Opto-Science and Technology (1998), the JSPS Prize (2007), and the Japan Academy Medal (2007). He is recognized for his pioneering work on ultrahigh-precision coherent control, where he utilized ultrashort laser pulses to visualize and control quantum interference in molecules on the picometer and femtosecond spatiotemporal scales. Recently he is interested in decoherence: how the quantum superposition created with an ultrashort optical pulse is destroyed by the interaction with an environment.