III-D Development of High-Precision Coherent Control and Its Applications

Coherent control is based on manipulation of quantum phases of wave functions. It is a basic scheme of controlling a variety of quantum systems from simple atoms to nanostructures with possible applications to novel quantum technologies such as bond-selective chemistry and quantum computation. Coherent control is thus currently one of the principal subjects of various fields of science and technology such as atomic and molecular physics, solid-state physics, quantum electronics, and information science and technology. One promising strategy to carry out coherent control is to use coherent light to modulate a matter wave with its optical phase. We have so far developed a high-precision wave-packet interferometry by stabilizing the relative quantum phase of the two molecular wave packets generated by a pair of femtosecond laser pulses on the attosecond time scale. We will apply our high-precision quantum interferometry to gas, liquid, solid, and surface systems to explore and control various quantum phenomena.

III-D-1 Visualizing Picometric Quantum Ripples of Ultrafast Wave-Packet Interference

KATSUKI, Hiroyuki[1]; CHIBA, Hisashi[1]; GIRARD, Bertrand[2]; MEIER, Christophe[2]; OHMORI, Kenji[1]
([1]IMS and JST/CREST; [2]Univ. Paul Sabatier (Toulouse III))

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Interference fringes in vibrating molecules are a signature of quantum mechanics, but are often so short-lived and closely spaced that they elude visualization. We have experimentally visualized dynamical quantum interferences, which appear and disappear in less than 100 femtoseconds in the iodine molecule synchronously with the periodic crossing of two counterpropagating nuclear wave packets. The obtained images have picometer and femtosecond spatiotemporal resolution, representing a detailed picture of the quantum interference.

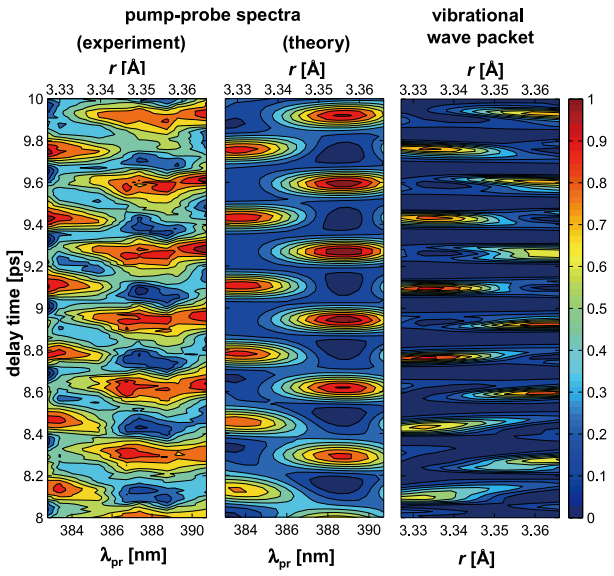

Figure 1. Contour plots of the experimental pump-probe signal (left), the simulated pump-probe signal (middle), and parts of the vibrational wave packet (right). The pump-probe spectra clearly reflect the spatiotemporal nodal structure of the wave-packet interferences.

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III-D-2 Real-Time Observation of Phase-Controlled Molecular Wave-Packet Interference

OHMORI, Kenji[1]; KATSUKI, Hiroyuki[1]; CHIBA, Hisashi[1]; HONDA, Masahiro[2,3]; HAGIHARA, Yusuke[2,3]; FUJIWARA, Katsutoshi[3]; SATO, Yukinori[2,3]; UEDA, Kiyoshi[2,3]
([1]IMS and JST/CREST; [2]JST/CREST; [3]Tohoku Univ.)

[*Phys. Rev. Lett*. **96**, 093002 (4 pages) (2006)]

The quantum interference of two molecular wave packets has been precisely controlled in the *B* electronic state of the I_2 molecule by using a pair of fs laser pulses whose relative phase is locked within the attosecond time scale and its real time evolution has been observed by another fs laser pulse. It is clearly observed that the temporal evolution changes drastically as a function of the relative phase between the locked pulses, allowing us to read both amplitude and phase information stored in the wave functions of the molecular ensemble.

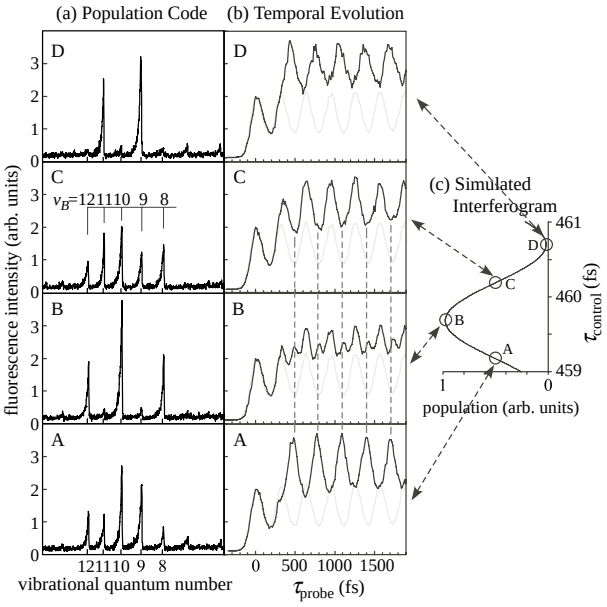

Figure 1. Wave packet interference measured and simulated with the pump and control delay $\tau_{control}$ tuned to ~1.5 T_{vib} (~460 fs), where T_{vib} is a classical vibrational period of I_2. (a) *E–B* excitation spectrum, which we call the "POPULATION CODE," measured by scanning the wavelength of the ns probe pulse delayed by ~35 ns from the pump and control pulses. The relative phase θ_{p-c} of the pump and control pulses is increased in steps of ~π/2 in going from code A to code D. (b) Real time evolutions of the wave-packet interference measured with the same θ_{p-c}'s as for codes A-D. The shaded trace is the evolution measured without the control pulse and is displayed for reference. Each trace is a summation of four repeated scans. The origin of the probe delay (τ_{probe} = 0) denotes a position of the top of the first undulation in each measured trace. The vertical scaling of each trace is arbitrary and is normalized by the height of its first undulation. (c) Simulation of $\tau_{control}$-dependence of the population of v_B = 10. The arrows stand for approximate correspondences in θ_{p-c}, and not in the absolute values of $\tau_{control}$.
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III-D-3 Implementation of Quantum Gate Operations in Molecules with Weak Laser Fields

TERANISHI, Yoshiaki[1]; OHTSUKI, Yukiyoshi[1]; HOSAKA, Kouichi[2]; CHIBA, Hisashi[2]; KATSUKI, Hiroyuki[2]; OHMORI, Kenji[2]
([1]Tohoku Univ. and JST/CREST; [2]IMS and JST/CREST)

[*J. Chem. Phys.* **124**, 114110 (9 pages) (2006)]

We numerically propose a way to perform quantum computations by combining an ensemble of molecular states and weak laser pulses. A logical input state is expressed as a superposition state (a wave packet) of molecular states, which is initially prepared by a designed femtosecond laser pulse. The free propagation of the wave packet for a specified time interval leads to the specified change in the relative phases among the molecular basis states, which corresponds to a computational result. The computational results are retrieved by means of quantum interferometry. Numerical tests are implemented in the vibrational states of the B state of I_2 employing controlled-NOT gate, and 2 and 3 qubits Fourier transforms. All the steps involved in the computational scheme, *i.e*., the initial preparation, gate operation, and detection steps, are achieved with extremely high precision.

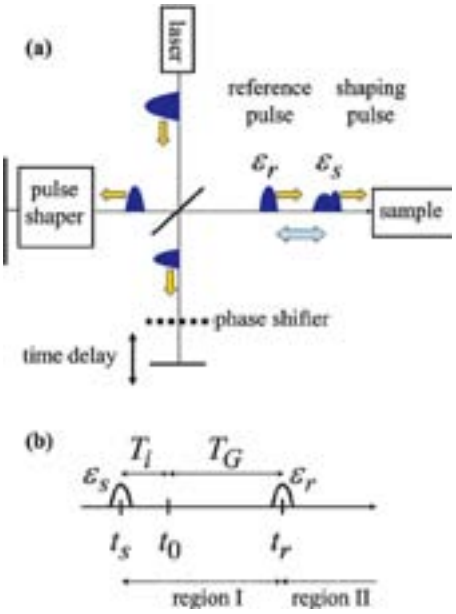

Figure 1. (a) Schematic of the experimental setup. (b) Pulse sequence for quantum computation. The times, t_s, t_0, and t_r, specify the temporal peak of the shaping pulse, the initial separation time, and the temporal peak of the reference pulse, respectively. The time intervals are referred to as the input preparation time $T_i = t_0 - t_s$ and the gate operation time $T_G = t_r - t_0$.
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