

Development of High-Precision Coherent Control and Its Applications

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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.

1. Actively Tailored Spatiotemporal Images of Quantum Interference on the Picometer and Femtosecond Scales¹⁾

Interference fringes of quantum waves weave highly regular space-time images, which could be seen in various wave systems such as wave packets in atoms and molecules, Bose Einstein condensates, and Fermions in a box potential. We have experimentally designed and visualized spatiotemporal images of dynamical quantum interferences of two counter-

propagating nuclear wave packets in the iodine molecule; the wave packets are generated with a pair of femtosecond laser pulses whose relative phase is locked within the attosecond time scale. The design of the image has picometer and femtosecond resolutions, and changes drastically as we change the relative phase of the laser pulses, providing a direct spatiotemporal control of quantum interferences.

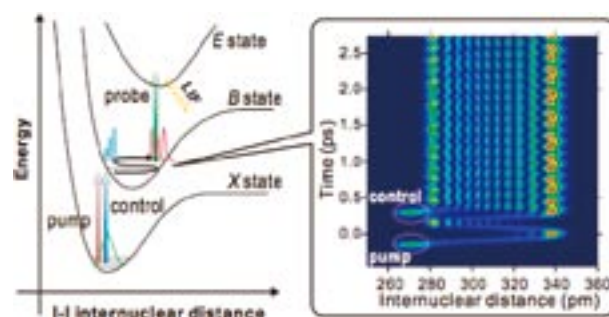


Figure 1. Pump-control-probe scheme for tailoring and visualizing the quantum carpet in the iodine molecule. (Left) Three femtosecond laser pulses are employed. Two of them are phase-locked and used as the pump and control pulses, and the other one is not phase-locked and is used as the probe pulse. The fluorescence signal induced by the probe pulse is measured with a photomultiplier tube attached to a monochromator. (Right) The model simulation of the quantum interference of two wave packets created on the *B*-state potential curve of the iodine molecule by the pump and control pulses. The interference shows a highly regular space-time image that looks like a carpet.

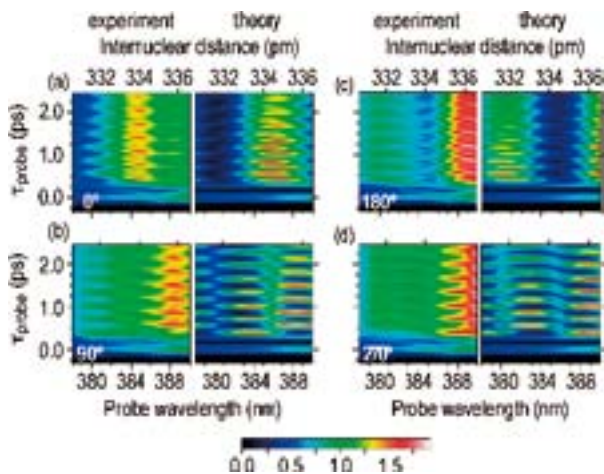


Figure 2. Comparisons of the quantum carpets measured (left) and simulated (right) at the pump-control relative phases θ_{pc} = (a) 0° , (b) 90° , (c) 180° , and (d) 270° . The color scaling is common within each set of measured or simulated carpets; the maxima of those two sets have the same color. The origin $\tau_{probe} = 0$ of the simulated carpet denotes a position of the top of the first oscillation around the outer turning point. The simulations include the interactions with the pump, control, and probe pulses. The parameter $\tau_{control}$, used for the simulation of $\theta_{pc} = 0^\circ$, was 468.920 fs.

2. Optical Control and Mode Selective Excitation of Coherent Phonons in $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$

Femtosecond time-resolved reflectivity measurement is performed on $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ films. Coherent phonons of both the Ba–O and Cu–O modes are observed at frequencies of 3.4 and 4.3 THz, respectively. Amplitudes of both Ba–O and Cu–O modes are optically manipulated by using a pair of femtosecond pulses, the separation time of which is controlled. Coherent phonons of the Ba–O and Cu–O modes are completely suppressed at the double-pulse separation times of 135.0 and 108.5 fs and those amplitudes are enhanced at 217 and 270 fs, respectively.

Award

KATSUKI, Hiroyuki; PCCP Prize.

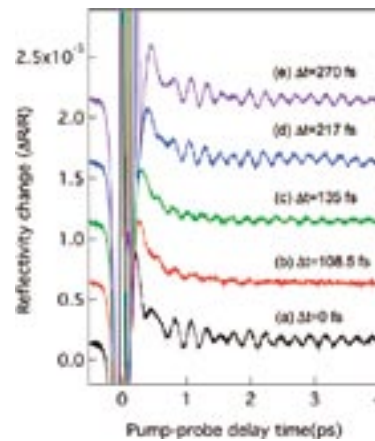


Figure 3. Time-resolved reflectivity of a photo-excited $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film by irradiation of a pair of femtosecond laser pulses at double-pulse separation time of 0 (a), 108.5 (b), 135 (c), 217 (d), and 270 fs (e).

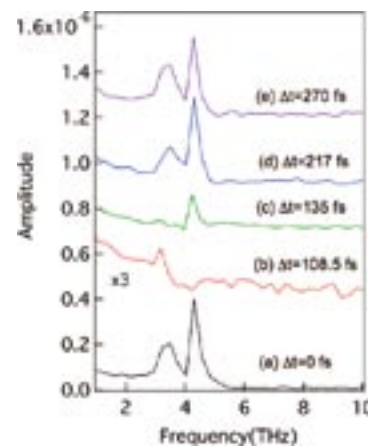


Figure 4. Fourier transformation of the transient reflectivity change of a photo-excited $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ thin film by irradiation of a pair of femtosecond laser pulses at double-pulse separation time of 0 (a), 108.5 (b), 135 (c), 217 (d), and 270 fs (e).

References

- 1) H. Katsuki, H. Chiba, C. Meier, B. Girard and K. Ohmori, *Phys. Rev. Lett.* **102**, 103602 (2009).
- 2) H. Takahashi, K. Kato, H. Nakano, M. Kitajima, K. Ohmori and K. G. Nakamura, *Solid State Commun.* in press.