

Development of High-Precision Coherent Control and Its Applications

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



OHMORI, Kenji
Professor



TAKEI, Nobuyuki
Assistant Professor

GOTO, Haruka
SOMMER, Christian
NAKAGAWA, Yoshihiro

Post-Doctoral Fellow
Post-Doctoral Fellow
Graduate Student

KOYASU, Kuniaki
INAGAKI, Itsuko
YAMAGAMI, Yukiko

Graduate Student
Secretary
Secretary

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 nano-structures 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 femto-second 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. Optically Engineered Quantum Interference of Delocalized Wavefunctions in a Bulk Solid: The Example of Solid Para-Hydrogen¹⁾

Local excitations of indistinguishable particles in a solid are quantum-mechanically superposed to give delocalized wave functions. Their interference is often so short-lived that it eludes observation and manipulation. Here we have actively controlled interference of delocalized vibrational wave func-

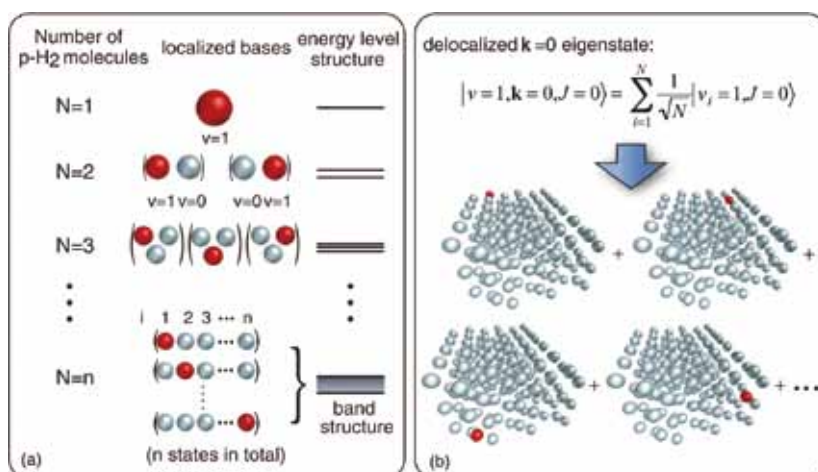


Figure 1. Formation of the delocalized vibron in solid p -H₂. (a) A band structure arises from the interaction among many indistinguishable p -H₂ molecules, one of which is vibrationally excited. The red and blue spheres stand for the vibrationally excited and ground-state p -H₂ molecules, respectively. (b) Every p -H₂ molecule within a single crystal has an equal probability to be vibrationally excited. These locally excited states $|v_i = 1\rangle$ ($i = 1, \dots, N$) are coherently superposed to give a delocalized vibron state $|v = 1, \mathbf{k} = 0\rangle$. This figure has been adopted from reference 1.

tions in solid *para*-hydrogen produced by a pair of ultrashort laser pulses. The ultrafast evolution of their interference changes from almost completely constructive (amplification by a factor of ~ 4) to destructive when we change the timing of those two laser pulses by only 4 fs. This active control serves as an experimental tool to investigate the spatiotemporal evolution of a wave function in a bulk solid.

2. Wavepacket Interferometry for Nuclear Densities and Flux Densities²⁾

The traditional wavepacket interferometry for nuclear densities is extended to nuclear flux densities. Accordingly, a molecule vibrating in an electronic excited state may be

prepared such that at a given time, the nuclear densities correspond to a broad distribution of the molecular bond, from short to long distances, which is subdivided into a chain of lobes. We discover that neighbouring lobes, or groups of lobes, may flow towards alternating directions, implying alternating bond stretches and bond compressions. The corresponding nuclear flux densities may be controlled by appropriate parameters of the two laser pulses, which generate the underlying interferences. Similar patterns of the nuclear densities and flux densities may also be created by a single laser pulse, which may cause interferences of the overlapping tail and head of a wavepacket as they run towards or away from a turning point, respectively. The phenomena are demonstrated for the model system $I_2(B)$.

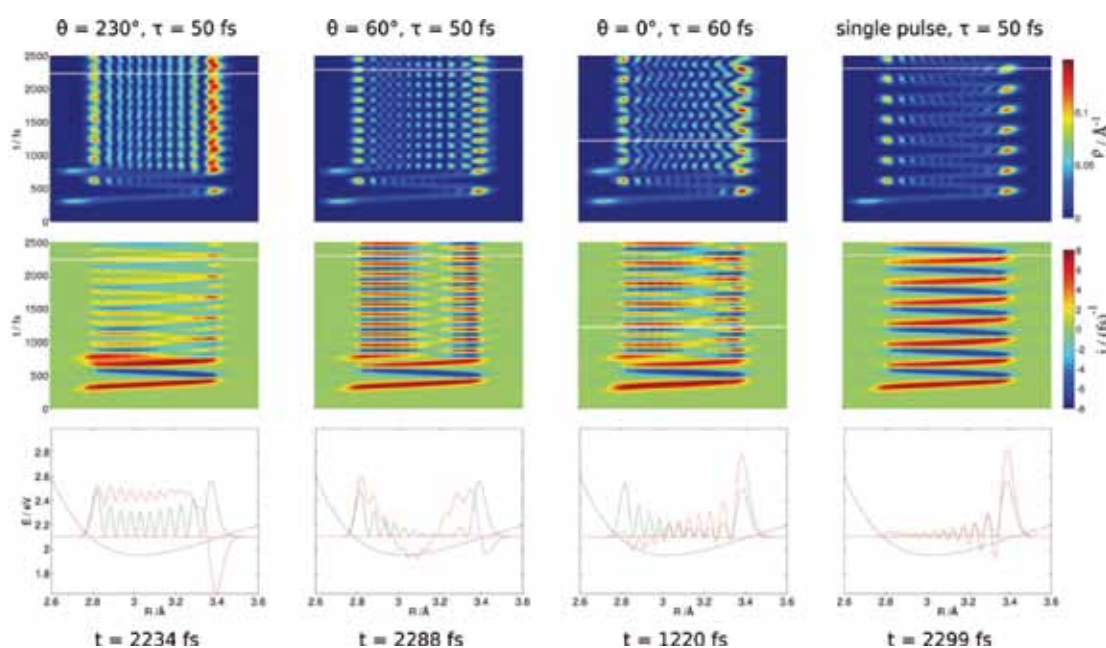


Figure 2. Wavepacket interferometry for the nuclear densities $\rho(R, t)$ (top panels) and flux densities $j(R, t)$ (middle panels) of the model $I_2(B)$, after excitations by two sub-pulses (first three columns) or by a single pulse (last column) versus nuclear bond length R (abscissa) and time t (ordinate). The values of $\rho(R, t)$ and $j(R, t)$ are indicated by colour codes on the right-hand side. The values of the control parameters of the laser sub-pulses (the duration τ and the phase θ of the second sub-pulse) are specified above the top panels. All other parameters of the laser pulses are fixed. The bottom panels show snapshots of $\rho(R, t)$ (green lines) and $j(R, t)$ (red lines) embedded in the PES of the electronic B -state (blue line) at specific times, which are indicated by horizontal lines in the top panels, and specified below the bottom panels. The new phenomena are discussed in detail in the text. This figure has been adopted from reference 2.

References

- 1) H. Katsuki, Y. Kayanuma and K. Ohmori, *Phys. Rev. B* **88**, 014507 (2013).
- 2) T. Bredtmann, H. Katsuki, J. Manz, K. Ohmori and C. Stemmler, *Mol. Phys.* **111**, 1691–1696 (2013). (invited paper).