III-F Development of High-Precision Coherent Control and Its Application

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 fs laser pulses in 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-F-1 Molecular Wave-Packet Interferometry with Attosecond Quantum Phase Manipulation

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Wave packet (WP) interferometry is a clear manifestation of wave nature of matter, and 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. The key technique in the WP interferometry is fine tuning of the delay between two light pulses that produce a pair of WP's with a precision far better than the quantum oscillation cycle of the WP's, say few femtoseconds (fs) to attoseconds (as). The WP's are then phase-locked and produce stable interference. We have constructed an "attosecond phase modulator (APM)": a device for tuning the delay τ between two UV fs pulses with attosecond stability and resolution. We have utilized this APM to create an unprecedented high-precision WP interferometer with a dilute ensemble of the HgAr van der Waals complex; our interferometer displayed almost 100% fringe contrast as a function of the delay τ between two UV fs pulses at 254 nm (→ Figure 1). Moreover we have demonstrated the dephasing and rephasing of the interferograms of consecutive vibrational eigenstates within WP's, which arise from a subtle difference in the quantum oscillation cycles of each eigenstates, different from the well-known collapse and revival of the electron WP's in atoms.[1)] Our high precision interferometer makes it possible to create virtually arbitrary relative superpositions of the three vibrational eigenstates within a WP only by tuning a single parameter τ. It is pointed out that the interference structure can be retrieved from the population information stored in the thermal ensemble of molecules even after the coherence is wiped out (→ Figure 2). All these features are quite general in WP interference and therefore provide basis for opening new perspective of coherent control in a wide variety of quantum systems.

Reference
1) John A. Yeazell, Mark Mallalieu and C. R. Stroud, Jr., *Phys. Rev. Lett.* **64**, 2007 (1990).

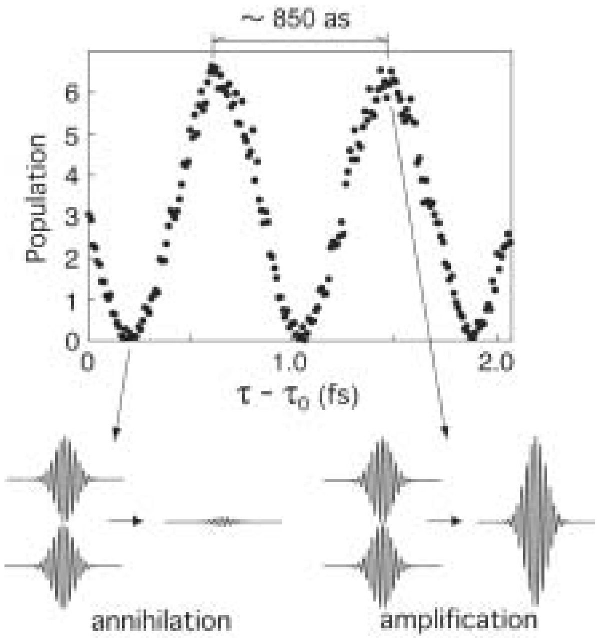

Figure 1. An example of the quantum interferograms of two molecular wave packets moving on the $A(^30^+)$-state potential curve of the Hg-Ar vdW complex. The interferogram displays almost 100% fringe contrast as a function of the inter-pulse delay τ tuned with attosecond stability and resolution. Its top and bottom represent amplification and annihilation of the wave packet.

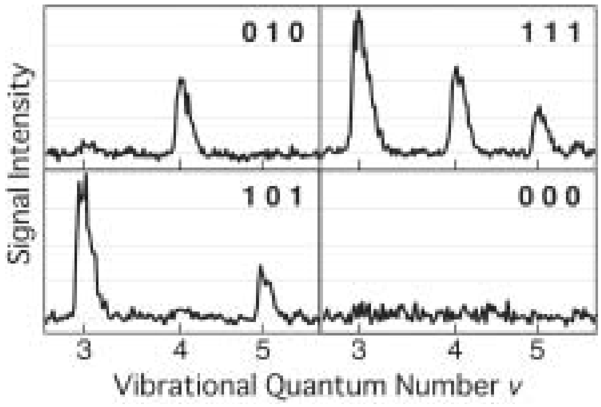

Figure 2. Population codes written in a Hg-Ar vdW complex by using wave-packet interference. Particular information can be encoded as a relative superposition of the vibrtional eigenstates within a wave packet, and that information can be retrieved as a population code stored in the ensemble of molecules even after the coherence is wiped out.