High-Precision Coherent Control of Molecular Wave Packets

HIROYUKI KATSUKI, Institute for Molecular Science, National Institutes of Natural Sciences, Japan, KOICHI HOSAKA, HISASHI CHIBA, KENJI OHMORI, INSTITUTE FOR MOLECULAR SCIENCE, NATIONAL INSTITUTES OF NATURAL SCIENCES, JAPAN TEAM, CREST, JAPAN SCIENCE AND TECHNOLOGY AGENCY, JAPAN COLLABORATION, THE GRADUATE UNIVERSITY FOR ADVANCED STUDIES (SOKENDAI), JAPAN COLLABORATION — The quantum interference of two vibrational wave packets has been precisely controlled in the electronically excited state of a diatomic molecule by using a pair of fs laser pulses whose relative phase \( \phi \) is locked within the attosecond time scale, and the real time evolution of that interference has been observed by another fs probe pulse. The real-time evolution shows a clear dependence on \( \phi \). We have also measured a population code, which is a population ratio among the vibrational eigenstates within a WP. The population code also shows a clear dependence on \( \phi \). The ordinary frequency domain interpretation based on the spectral interference of locked pulses may be useful to elucidate \( \phi \) dependence of population codes, but is no longer suitable for the present real-time observation. The combination of a population code and real-time evolution is useful to obtain both phase and amplitude information stored in a WP.

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