IMAGING ROTATIONAL WAVE PACKETS IN MOLECULES AND CLUSTERS

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Irradiation of ultrashort laser pulses onto a molecular ensemble inherently results in the creation of a quantum wave packet (WP), i.e., a coherent superposition of several (or sometimes many) eigenstates. It is an ultimate goal in modern molecular physics to establish the way for creating WPs as designed, and it is also essential for advanced WP manipulation to know how to characterize the WP experimentally.

Over the last two decades, intense nonresonant excitation has been extensively adopted to realize "nonadiabatic molecular alignment" or "nonadiabatic rotational excitation," where the impulsive torque due to anisotropic molecular polarizability instantaneously aligns the molecules and their rotation is coherently excited in the vibronic ground-state manifold.[a] Evolution of the rotational WP thus created can be tracked as a series of images for the time-dependent molecular orientational distribution, e.g., by implementing pump-probe Coulomb explosion ion-imaging measurements. We will represent some examples of such "molecular movies" taken with a newly developed imaging configuration,[b] which is capable to clearly capture the time-dependent nodal structures, instantaneous alignment, angular dispersion, and fractional revivals of the rotational WP while the molecular ensemble keeps rotating in one direction.[c]

Wave-packet imaging will also be developed as a new approach in molecular spectroscopy, since energy intervals between the eigenstates that constitute the WP are encoded in the molecular movies shoot by the method. We will show some recent results along this direction; rotational spectra have been successfully extracted for homodimers of nonpolar molecules, which have been scarcely investigated because of the difficulty in recording or analyzing their microwave or infrared spectrum.

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[a] Y. Ohshima and Hasegawa, Int. Rev. Chem. Phys. **29**, 619 (2010).

[b] K. Mizuse, R. Fujimoto, and Y. Ohshima, Rev. Sci. Instrum. **90**, 103107 (2019).

[c] K. Mizuse, K. Kitano, H. Hasegawa, and Y. Ohshima, Sci. Adv. **1**, e1400185 (2015).