

Supplementary Materials for Quantum unidirectional rotation directly imaged with molecules

Kenta Mizuse, Kenta Kitano, Hirokazu Hasegawa, Yasuhiro Ohshima

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The PDF file includes:

Fig. S1. Fourier transformation analyses of $\langle P_n(\cos\phi) \rangle$ for the higher-intensity experiment.

Fig. S2. Time-dependent signal intensity as a measure of the probability in the xy plane.

Fig. S3. Fourier transformation analyses of $\langle P_n(\cos\phi) \rangle$ for the lower-intensity experiment.

Legends for movies S1 to S3

Other Supplementary Material for this manuscript includes the following:

(available at www.advances.sciencemag.org/cgi/content/full/1/6/e1400185/DC1)

Movie S1 (.mp4 format). Unidirectional rotation induced by higher-intensity pulses (all frames of Fig. 3).

Movie S2 (.mp4 format). Calculated unidirectional molecular rotation, corresponding to movie S1.

Movie S3 (.mp4 format). Experimental movie for the lower-intensity case.

Supplementary Materials

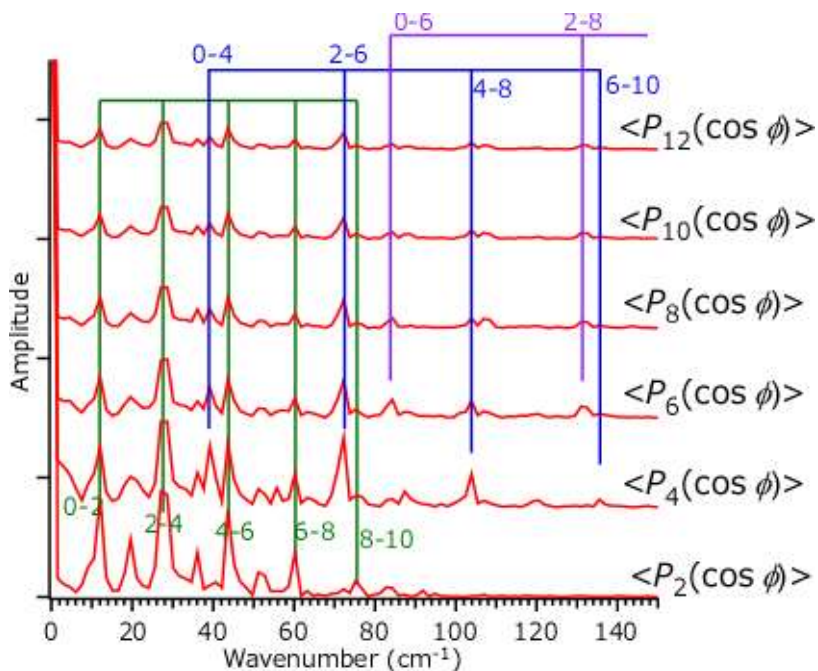


Fig. S1. Fourier transformation analyses of $\langle P_n(\cos \phi) \rangle$ for the higher-intensity experiment.

The label J_1 – J_2 indicates the coherence between the displayed two states. This figure suggests almost negligible contribution of $J \geq 10$ in the wave packet dynamics. Labels are shown only for even J for simplicity.

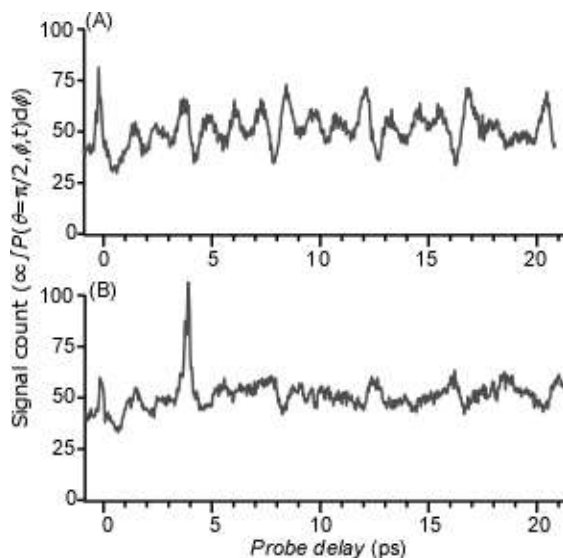


Fig. S2. Time-dependent signal intensity as a measure of the probability in the xy plane.

(A) For the single pump experiment, as in Fig. 2. (B) For the case of the double pump experiment, as in Figs. 3 and 4.

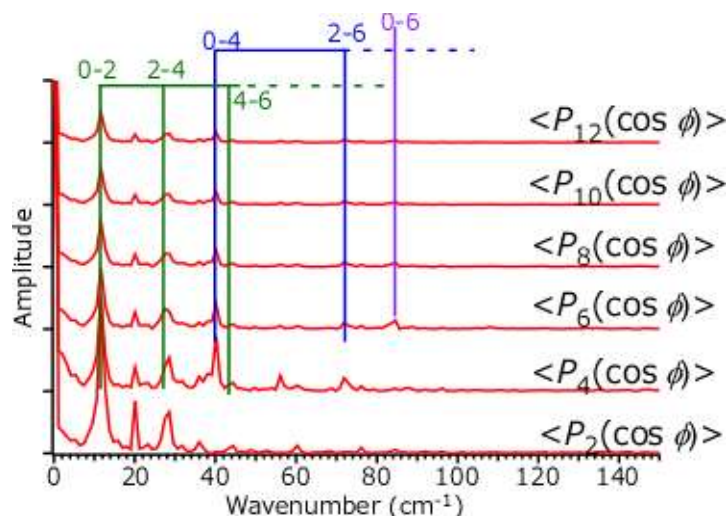


Fig. S3. Fourier transformation analyses of $\langle P_n(\cos\phi) \rangle$ for the lower-intensity experiment. This figure suggests almost negligible contribution of $J \geq 6$ in the wave packet dynamics. Labels are shown only for even J for simplicity.

Movie S1. Unidirectional rotation induced by higher-intensity pulses (all frames of Fig. 3). In each frame, the left picture shows the observed N^{3+} ion image (after the calibration). The double ring structure comes from the two channels of Coulomb explosion (N^{3+} from N_2^{4+} and N_2^{5+}), in which higher charge states lead to larger kinetic energy release (outer ring). The right one is the polar plots of the observed angular probability. The polarizations of the pulses are shown as green arrows at around $t = 0$ and 4 ps. We note the pump-probe overlap and imperfect circular polarization of the probe somewhat affect the shape of distributions at $t \leq 1$ ps.

Movie S2. Calculated unidirectional molecular rotation, corresponding to movie S1. As in Fig. 4, calculated three dimensional probability and corresponding projections are shown. See Fig. 4 and text for details of simulation.

Movie S3. Experimental movie for the lower-intensity case. All frames correspond to the full version of the plots in Fig. 5. We note the pump-probe overlap and imperfect circular polarization of the probe somewhat affect the shape of distributions at $t \leq 1$ ps.