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**Coherent Control of Single Molecules at Room Temp** NIEK VAN HULST, DAAN BRINKS, RICHARD HILDNER, ICFO - the Institute of Photonic Sciences (Barcelona) Spain — Electronic coherence plays a key role in natural processes like ultrafast energy transfer and charge separation. Coherent control has proven powerful, however in complex biosystems with different conformations and environments, the intrinsic inhomogeneity of the synchronized subset severely limits the achievable degree of control. The ultimate solution to overcome intrinsic inhomogeneities is the investigation of the behavior of one molecule at a time. Here we report the observation and manipulation of vibrational wave-packet interference and electronic coherence in *individual molecules* at ambient conditions. Adapting time and phase distribution of the optical excitation field to the dynamics of each molecule we achieve a superior degree of control. The time-phase maps show distinct diversity between different, yet chemically identical, molecules. We induce Rabi-oscillations and control the coherent superposition state in a single molecule. Broadly distributed coherence decay times are found for different individual molecules giving direct insight into the structural heterogeneity of the local surroundings. Our approach allows single-molecule coherent control in a variety of complex inhomogeneous systems and thus to study the role of coherence in energy transfer of single biocomplexes under natural conditions. D.Brinks *et al. Nature* **465**, 905 (2010); R.Hildner *et al. Nat.Physics* doi:10.1038/nphys1858 (2010).

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