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Ultrafast vibrational decoherence probed in the infrared near-field of molecular bonds MATTHIAS RANG, Forschungsinstitut am Goetheanum, Dornach, CH, XIAOJI G. XU, ANDREW C. JONES, MARKUS B. RASCHKE, University of Washington, Seattle — The strong intra- and intermolecular coupling in molecular systems is responsible for the dominance of the non-radiative over the radiative pathway in the ultrafast decoherence of vibrational excitations. The traditional coherent vibrational spectroscopy techniques therefore rely on nonlinear wavemixing to probe the underlying dynamics. Here we demonstrate the enhanced radiative far-field coupling from the optical near-field of vibrational excitations in block-copolymer nanostructures chosen as model system, by scattering scanning near-field optical microscopy (s-SNOM). The modified tip-scattered free-induction decay of the coherently excited vibrational mode following a broadband femtosecond infrared excitation is temporally resolved by interferometric homodyne detection. The results provide insight into spatial coherence and inhomogeneity with nanometer spatial resolution.

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