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Decoherence in quantum control: the role of the solvent

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Coherent control holds the promise of becoming a powerful spectroscopic tool for the study of complex molecular systems. Achieving control requires coherence in the quantum system under study. In the condensed phase, coherence is typically lost rapidly due to fluctuating interactions between the solvated molecule and its surrounding environment. Surprisingly, it is still possible to achieve a high degree of control. Here we investigate the degree of attainable control on a dye molecule when the fluctuations of its environment are systematically varied. A single successful learning curve for optimizing stimulated emission from the dye molecule in solution is reapplied for a range of solvents with varying viscosity. The optimization yields reveal a striking trend that is correlated directly to the dephasing time by a visco-elastic model. Our results provide clear evidence that the fluctuations of the environment limit the leverage of control on the molecular system.