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Ultrafast Dynamics of Vibration-Cavity Polariton Modes JEFF OWRUTSKY, ADAM DUNKELBERGER, KENAN FEARS, BLAKE SIMPKINS, Chemistry Division, Naval Research Laboratory, BRYAN SPANN, National Institute of Standards and Technology — Vibrational modes of polymers, liquids, and solvated compounds can couple to Fabry-Perot optical cavity modes, creating vibration-cavity polariton modes whose energy tunes with the cavity length and incidence angle. Here we report the pump-probe infrared spectroscopy of vibration-cavity polaritons in cavity-coupled $W(CO)_6$. At very early times, we observe quantum beating between the two polariton states find an anomalously low degree of excitation. After the quantum beating, we directly observe spectroscopic signatures of excited-state absorption from both polariton modes and uncoupled reservoir modes. An analytical expression for cavity transmission reproduces these signatures. The upper polariton mode relaxes ten times more quickly than the uncoupled vibrational mode and the polariton lifetime depends on the angle of incidence of the infrared pulses. Coupling to an optical cavity gives a means of control of the lifetime of vibration-cavity polaritons and could have important implications for chemical reactivity in vibrationally excited molecules.

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