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Coherent Raman spectroscopy with incoherent laser pulses XIAOJI XU, STANISLAV KONOROV, JOHN HEPBURN, VALERY MILNER — Contrary to the common belief that noise and decoherence are detrimental to spectroscopic measurements, we propose and experimentally demonstrate a new method of coherent Raman spectroscopy with spectrally broad incoherent laser pulses. Laser induced molecular vibrations are probed by broadband laser pulses with intentionally introduced spectral phase noise. The vibrational resonances are identified through intensity correlations in the noisy spectrum of the scattered anti-Stokes photons. Spectral resolution is neither limited by the pulse bandwidth nor sensitive to the temporal profile quality of the pulses. The method does not require complicated pulse-shaping setups, spectral multiplexing or spatial beam arrangements. It enables full utilization of the broad bandwidth of femtosecond pulses, and quick scanless retrieval of the vibrational beating frequencies.

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