

Spectroscopy in the Fourier Domain: Correlated Measurement of Rotational and Electronic Structure

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Femtosecond spectroscopy is used extensively to resolve ultrafast reaction mechanisms. Due to Heisenberg's uncertainty principle, the spectroscopic resolution of such experiments is low and rarely contains enough information to resolve molecular structure. As a result, femtosecond experiments are limited to clean samples with established molecular structure. In a novel experimental scheme, we combine short-pulse experiments with high-resolution rotational spectroscopy and obtain correlated structural and dynamic information, thereby breaking the limitations discussed above. We demonstrate the power of our approach with first experimental results for CS₂.