DEPARTMENT OF
Chemistry
UNIVERSITY OF WISCONSIN–MADISON

Ph.D. Dissertation Defense

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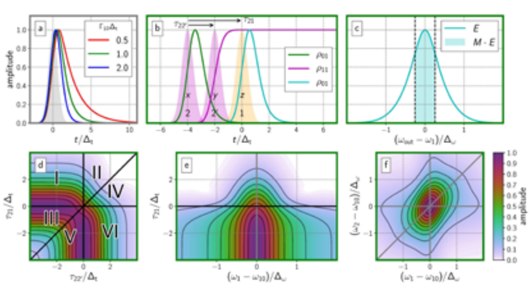
"Development of Frequency Domain Multidimensional Spectroscopy with Applications in Semiconductor Photophysics"

Coherent multidimensional spectroscopy (CMDS) encompasses a family of experimental strategies involving the nonlinear interaction between electric fields and a material under investigation. This approach has several unique capabilities: (1) resolving congested states, (2) extracting spectra that would otherwise be selection-rule disallowed, (3) resolving fully coherent dynamics, (4) measuring coupling, and (5) resolving ultrafast dynamics.

Frequency domain "Multi-resonant" CMDS (MR-CMDS) requires pulsed ultrafast light sources with tunable output frequencies. These pulses are directed into a material under investgation. The pulses interact with the material, and due to the specific interference between the multiple fields, the material is driven to emit a new pulse: the MR-CMDS signal. The MR-CMDS experiment involves tracking the intensity of this output signal as a function of different properties of the excitation pulses.

Because MR-CMDS is a family of related-but-separate experiments, each of them a multidimensional space, there are special challenges that must be addressed when designing a general-purpose MR-CMDS instrument. I present five strategies used to improve MR-CMDS: (1) processing software, (2) acquisition software (3) active artifact correction, (4) automated OPA calibration, and (5) finite pulse accountancy. Results in three material systems are presented heterostructure flakes.



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