Development of Frequency-Domain Multidimensional Spectroscopy Blaise Thompson

Tunability

Acquisition Extensibility Queue

Artifacts

Processing Conclusion

Supplement



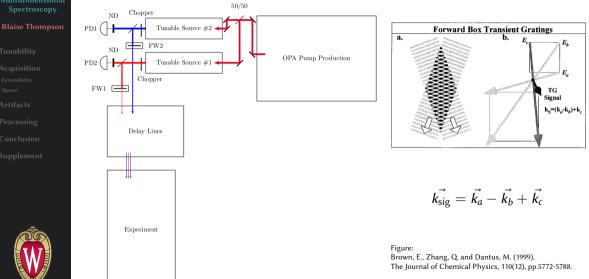
Development of Frequency-Domain Multidimensional Spectroscopy –Beyond Two Dimensions–

Blaise Thompson

University of Wisconsin-Madison

2018-04-23

Introduction to CMDS



Development of Frequency-Domain Multidimensional

Introduction to CMDS

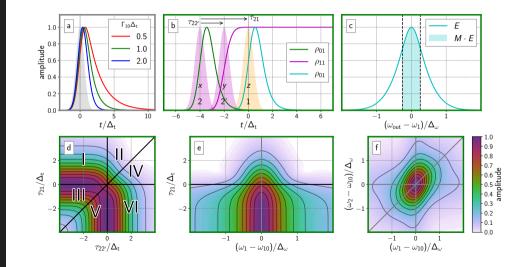
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Different phase matching conditions...

- transient grating $\vec{k_a} \vec{k_b} + \vec{k_c}$
- transient absorption
- DOVE

But also different color combinations and dimensions explored.



Pipeline

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What does the "pipeline" of MR-CMDS data acquisition and processing look like in the Wright Group?



How to increase data throughput and quality, while decreasing frustration of experimentalists?

MR-CMDS development

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[SUMMARY SLIDE FOR REMAINDER OF PRESENTATION]

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Control and Calibration of Optical Parametric Amplifiers

Two strategies for CMDS

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Two strategies for collecting multidimensional spectra:

Time Domain

- broadband pulses
- resolve spectra interferometrically
- ▶ fast (even single shot)
- robust

Frequency Domain

- narrowband pulses
- resolve spectra by tuning OPAs directly
- slow (lots of motor motion)
- fragile

Two strategies for CMDS

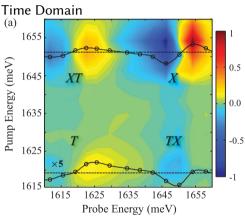
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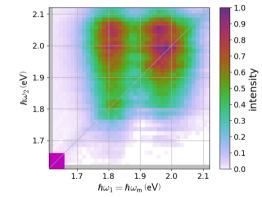
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Frequency Domain



Figure

Singh, A., Moody, G., Wu, S., Wu, Y., Ghimire, N., Yan, J., Mandrus, D., Xu, X. and Li, X. (2014). Coherent Electronic Coupling in Atomically Thin MoSe₂. Physical Review Letters, 112(21).

More bandwidth. Crucial for electronic states, band structure.

Bandwidth

A lot more bandwidth... through the usage of OPAs

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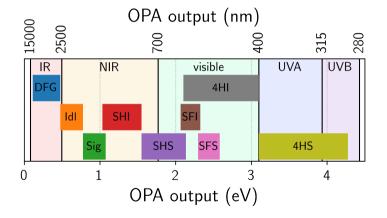
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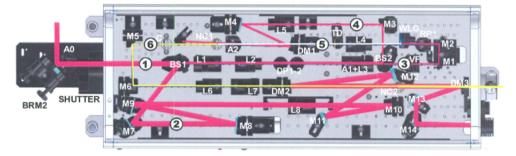


TOPAS-C

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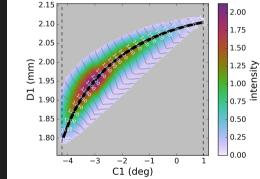
Two "stages", each with two motorized optics.

Automation

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Fully automated OPA tuning

- less than 1 hour per OPA
- can be scheduled for odd times
- high quality from global analysis
- reproducible
- unambiguous representations automatically generated

Other calibration steps also automated.



Acquisition

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Control of the MR-CMDS Instrument

The instrument

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Many kinds of component hardware

- monochromators
- delay stages
- filters
- OPAs
- \sim 10 settable devices, \sim 25 motors, multiple detectors.



Acquisition

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Postino

Position

Position

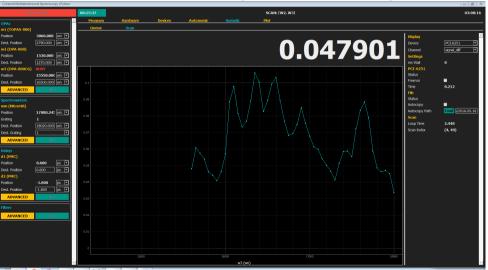
d1 (PMC)

d2 (PMC) Position

Acquisition



PyCMDS-unified software for controlling hardware and collecting data.



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Acquisition

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Processing Conclusion Supplement At its core, PyCMDS does something very simple...

Set, wait, read, wait, repeat.

Everything is multi-threaded (simultaneous motion, simultaneous read).
▶ decrease scan time by up to ~ 2×, more for complex experiments



Extensibility

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Shutter	
Dest. Position	14000.000 wn 🔽
Dest. Shutter	
w2 (TOPAS-C)	
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A modular hardware system that can be easily added to

- When a new OPA was installed on the picosecond system, PyCMDS was back in action the next day.
- Darien inherited a new delay stage from the Crim Group, and added it to PyCMDS in less than two days.
- New kinds of hardware also possible to add, although this is more difficult.

just need to copy a script and modify...

Development of Frequency-Domain Multidimensional Spectroscopy

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Multidimensional

- Tunability Acquisition Extensibility Queue
- Artifacts
- Processing Conclusion Supplement



This strategy can be incredibly productive!

Soon after the queue was first implemented, we collected more pixels in two weeks than had been collected over the previous three years. Development of Frequency-Domain Multidimensional Spectroscopy Blaise Thompson

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Acquisition

Artifact Rejection

Shots Processing

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[DIGITAL SHOTS PROCESSING—NO MORE BOXCARS]

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Processing

Processing

Data Processing



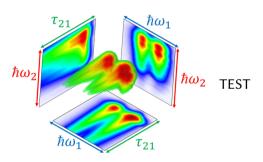
Dimensionality

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Flexible data model

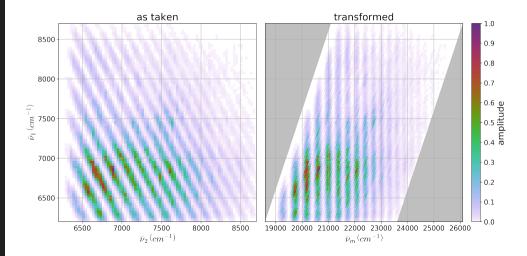
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Flexibility to transform into any desired "projection" on component variables.



Universal format

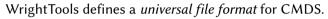
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- store multiple multidimensional arrays
- metadata

Import data from a variety of sources.

- previous Wright Group acquisition software
- commercial instruments (JASCO, Shimadzu, Ocean Optics)



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[CONCLUSION]

Conclusion

Acknowledgments

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- Conclusion

Wright Group

- Kyle Sunden
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- Skye Kain
- ► John
- (and more...)

Committee

- Kyoung-Shin Choi
- Randall Goldsmith
- Tim Bertram

UW-Madison Chemistry Department

- Rob McClain
- Pam Doolittle
- Bill Goebel
- Steve Myers

You, the audience! Questions?

Tuning

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Tuning curves—recorded correspondence between motor positions and output color.

Exquisite sensitivity to alignment and lab conditions—tuning required roughly once a week.

Manual tuning is difficult...

- high dimensional motor space
- difficult to asses overall quality
- several hours of work per OPA (sometimes, an entire day for one OPA)

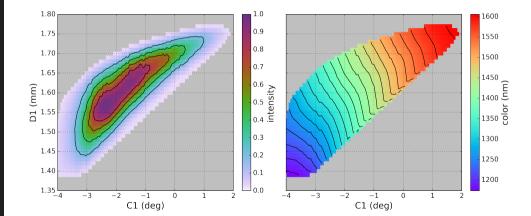
Preamp

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Modular hardware model

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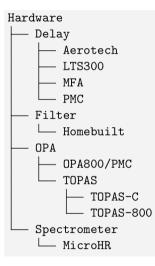
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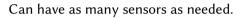




Modular sensor model

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Each sensor contributes one or more channels.

Sensors with size contribute new variables (dimensions).



Domains of CMDS

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CMDS can be collected in two domains:

- time domain
- frequency domain



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Multiple broadband pulses are scanned in *time* to collect a multidimensional interferogram (analogous to FTIR, NMR).

A local oscillator must be used to measure the *phase* of the output.

This technique is...

- ► fast (even single shot)
- robust

pulse shapers have made time-domain CMDS (2DIR) almost routine.

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In the Wright Group, we focus on *frequency* domain "Multi-Resonant" (MR)-CMDS.

Automated Optical Parametric Amplifiers (OPAs) are used to produce relatively narrow-band pulses. Multidimensional spectra are collected "directly" by scanning OPAs against each-other.

This strategy is...

- slow (must directly visit each pixel)
- fragile (many crucial moving pieces)

but! It is incredibly flexible.

Selection rules

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MR-CMDS can easily collect data without an external local oscillator.

This means... [BOYLE]

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[FIGURES FROM DAN'S PAPER]

Mixed domain