# A Robust, Fully Automated Algorithm to Collect High Quality OPA Tuning Curves

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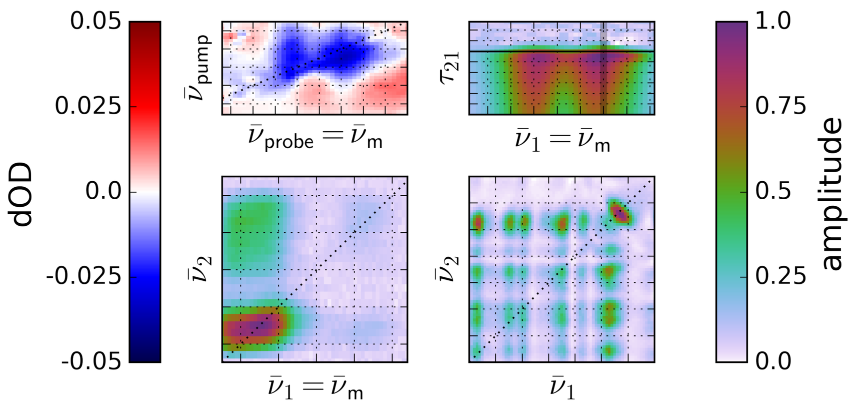
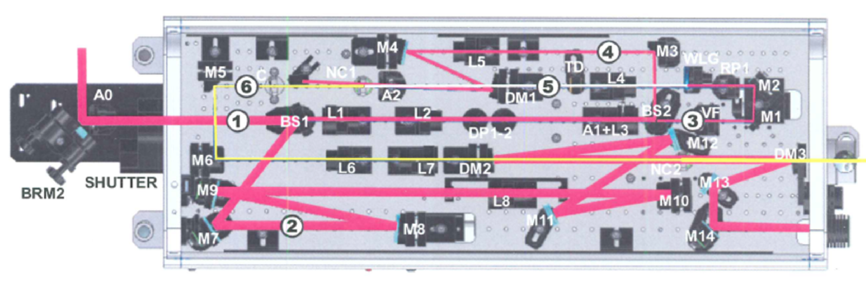
## Frequency-Domain CMDS

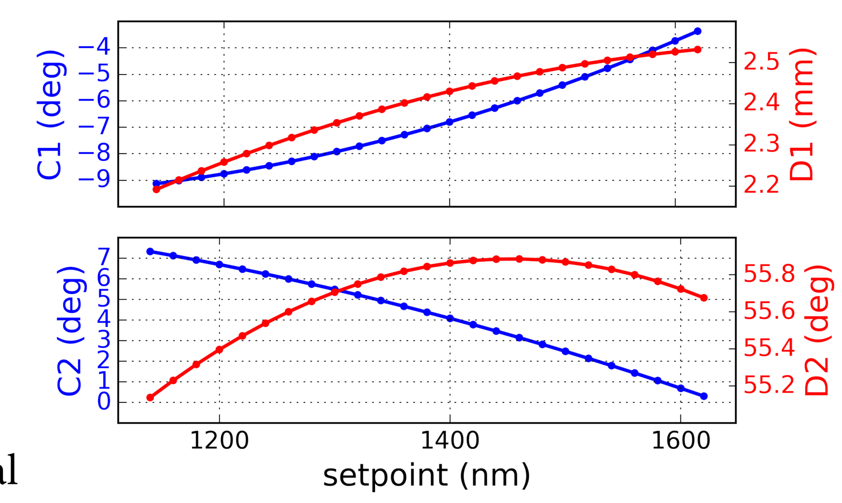



In frequency-domain CMDS, automated OPAs are used to actively scan excitation color axes. Four recent examples of frequency-domain CMDS in the Wright Group are shown to the left[1,2]. Each of these examples contains at least one scanned OPA axis.

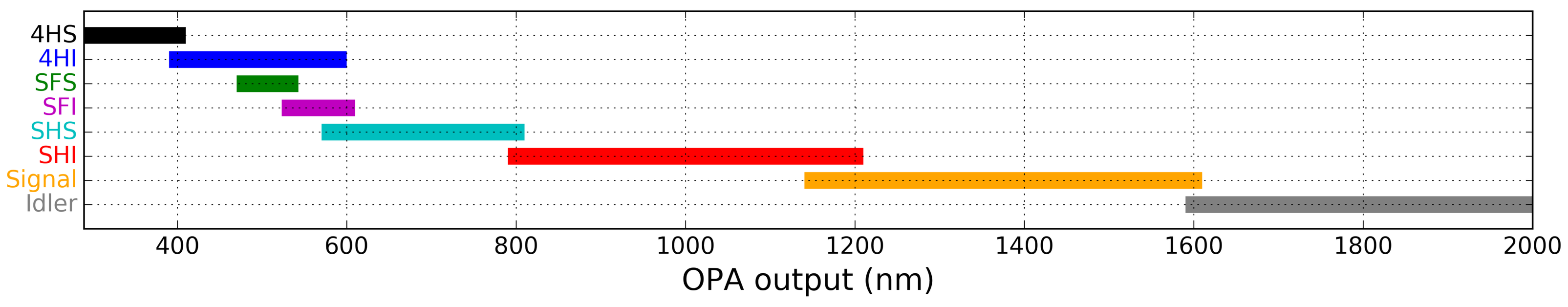
To accomplish these experiments, **exquisite OPA performance** is required. During the experiment, motors inside the OPA move to pre-recorded positions to optimize output at the desired color.




Adapted from Light Conversion TOPAS-C manual




Parametric conversion ('mixing') strategies are now readily available, extending 800 nm pumped OPA tuning range into the visible/near-IR (shown below) and mid-IR (not shown).




## Automated Tuning

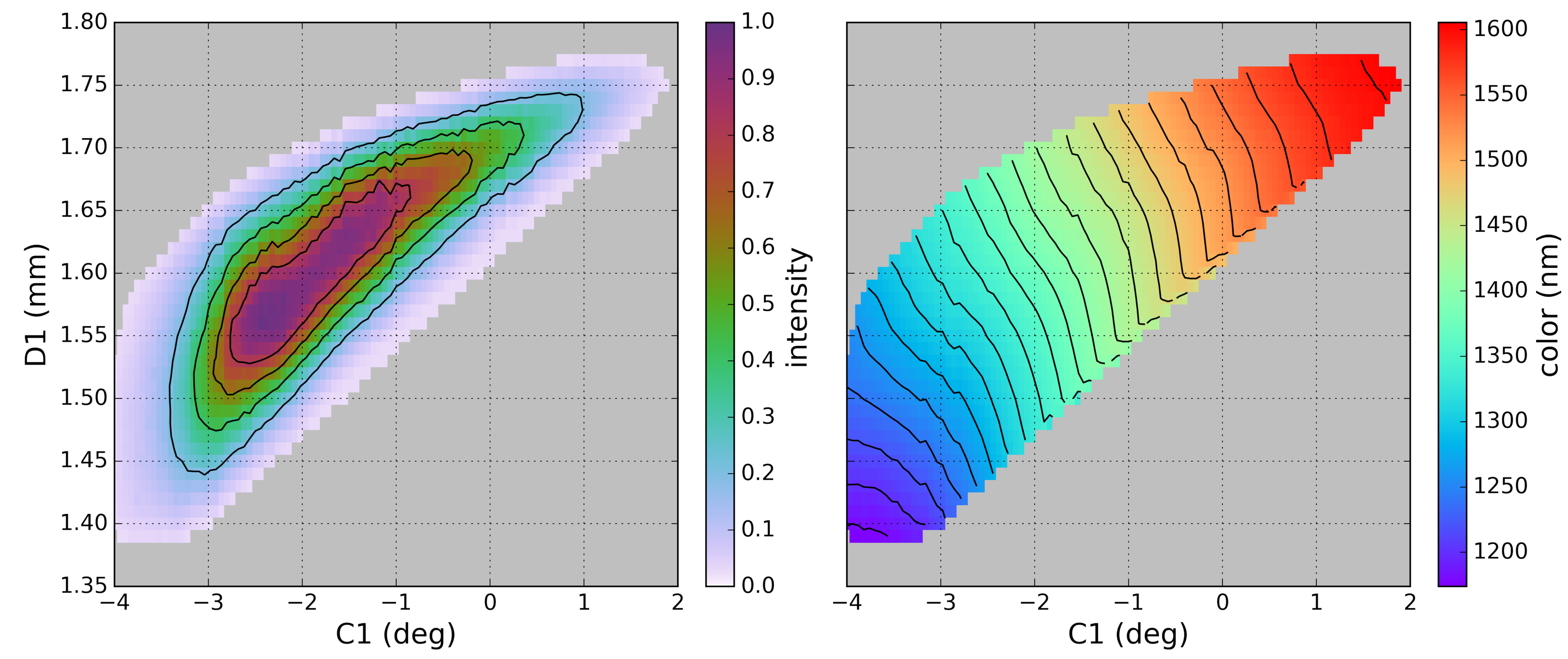
OPAs are very sensitive to changes in upstream lasers and lab conditions, so OPA tuning is regularly required. Manual OPA tuning can easily take a **full day**. Automated OPA tuning makes OPA upkeep **easier, faster,** and **more reproducible,** facilitating frequency domain experiments. The major challenges in automated OPA tuning, and our solutions, are below.

| | |
|---|---|
| 1. Expensive to take high resolution data | 1. Fitting and interpolation is robust to noise and step size |
| 2. Need smooth curves for interpolation, especially at edges where output is low | 2. Force curves through smoothing spline |
| 3. Optimization metrics are not necessarily separable along motor dimensions | 3. Multidimensional or iterative tuning procedures |

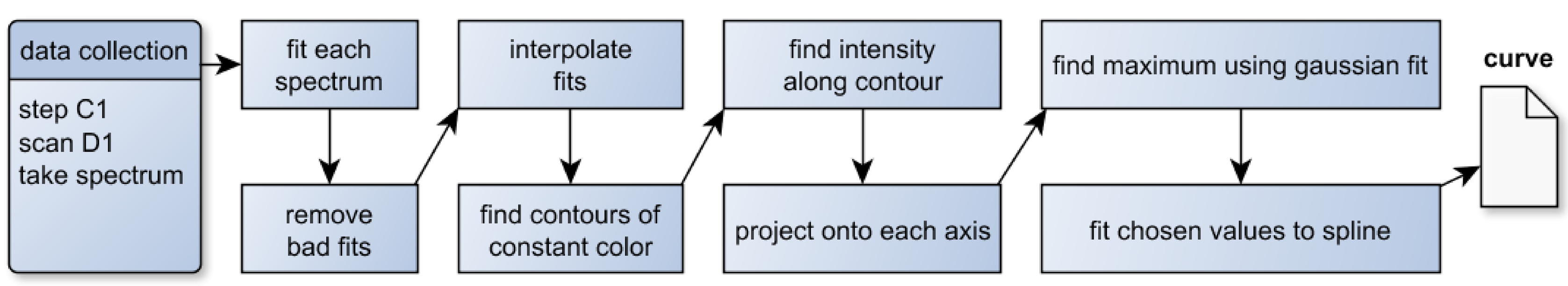
In the following we describe the specific implementation for ~35 fs Light Conversion model TOPAS-C OPAs.

## Preamp

In TOPAS-C OPAs, a small portion of input light is used to generate a signal seed in a BBO crystal `C1'. A motorized delay stage 'D1' is used to temporally overlap a particular color in chirped white light with 800 nm pump. C1 angle is tuned to optimize phase matching. Measured seed intensity and color for all combinations of C1 and D1 position are shown below.

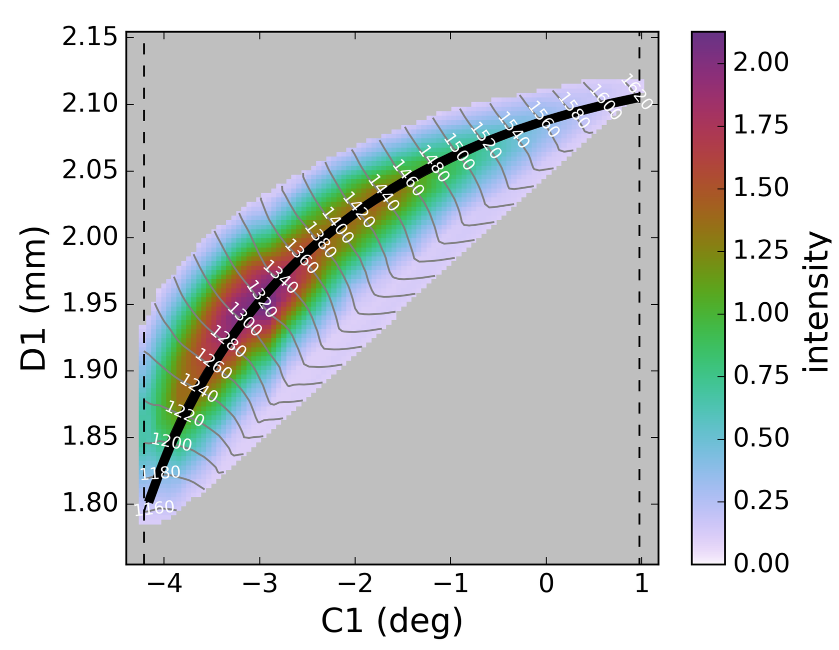



Output color and intensity are not separable along the preamp motor axes. We therefore use a multidimensional fitting strategy to find the best preamp motor positions, as shown below.



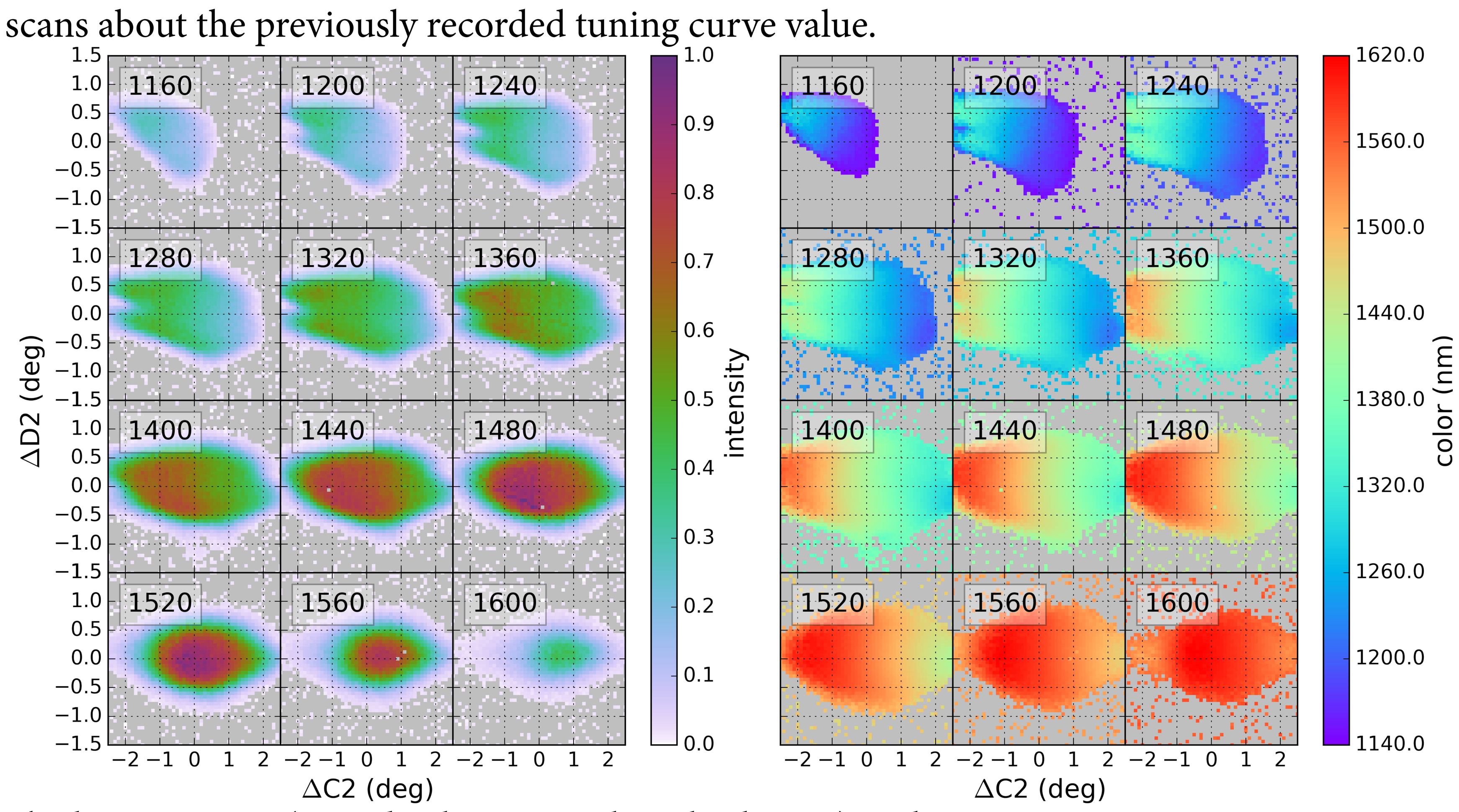

A representative preamp tune procedure output image is shown to the right. The thick black line is the final output curve. The dark grey lines are the contours of constant color. The colorbar shows the Delaunay-interpolated intensity values for each motor position.

Preamp tuning takes **less than 20 minutes**, in large part due to a NIR array detector which collects the full spectrum at each motor position.

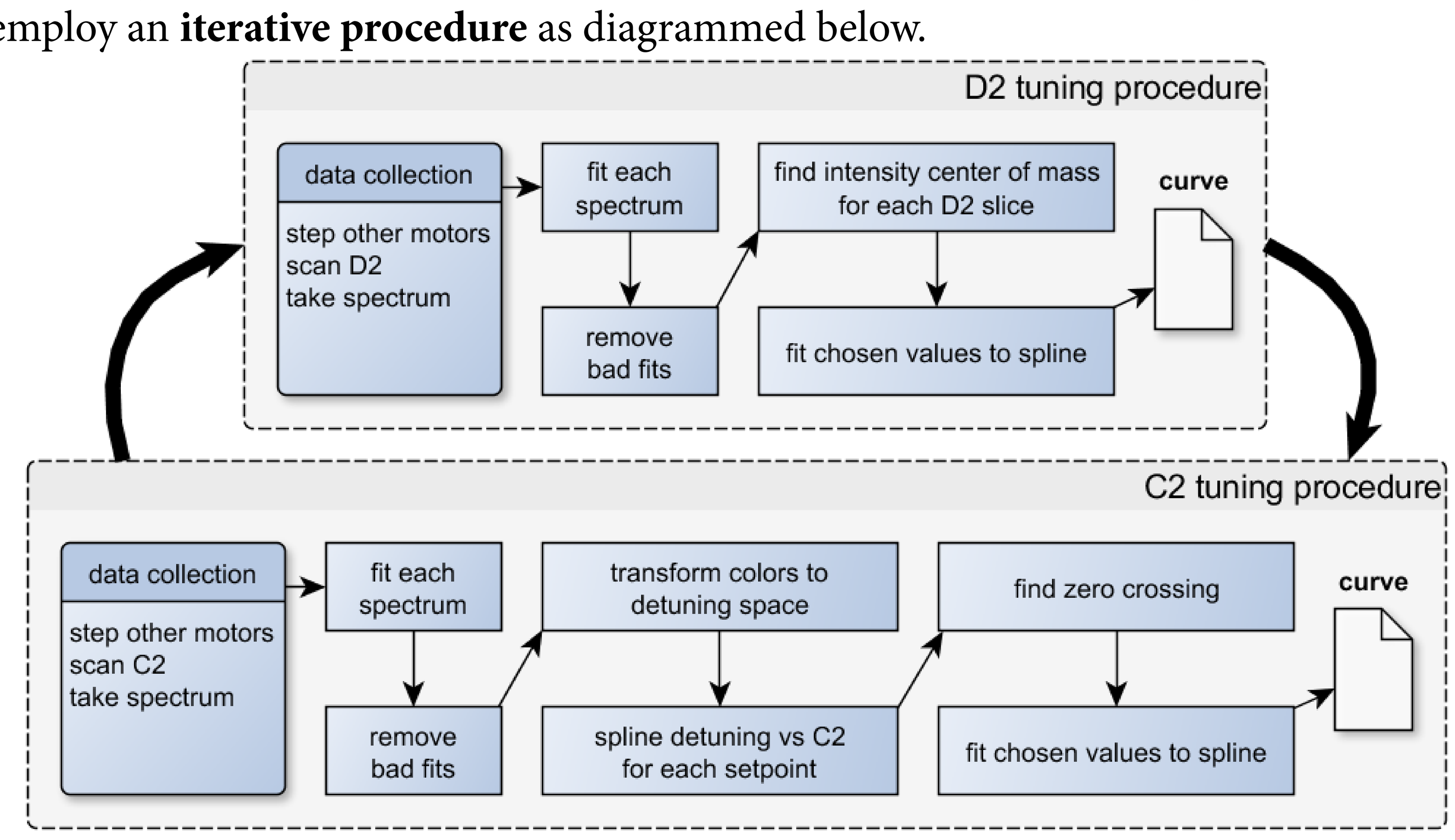



## Poweramp

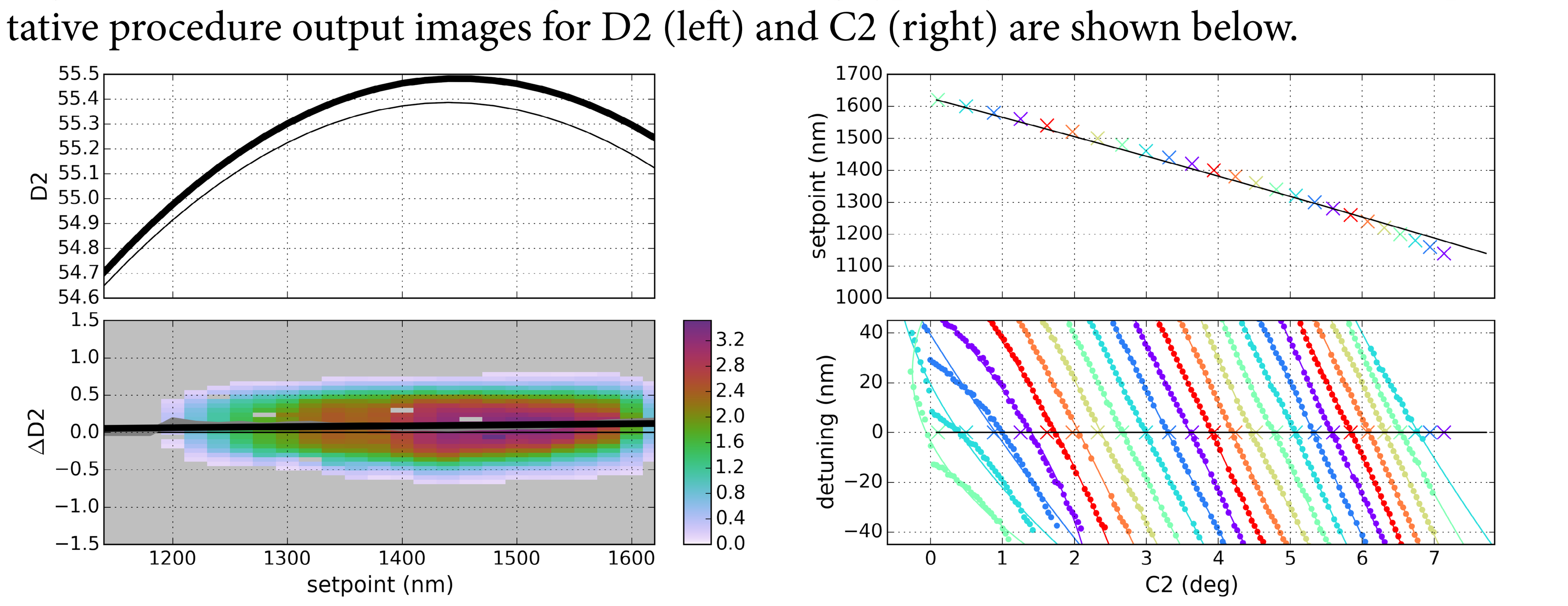
Once generated, the seed goes on to be amplified in a second BBO crystal 'C2' with the rest of the 800 pump. Optimizing this amplification step is primarily a matter of setting C2 angle. A small delay correction 'D2' is necessary to account for dispersion in the seed optics. To fully explore poweramp behavior, we need to take a C2-D2 scan for each seed color. Measured output intensity and color in this 3D space is represented below. Note that the motor axes are scans about the previously recorded tuning curve value.




The best position (zero displacement along both axes) is chosen to maximize output intensity while keeping the output color identical to the seed color. **Optimizing for zero detuning** rather than simply for output intensity has led to **better OPA performance** and stability. Like in the preamp case, color and intensity are not fully separable along the poweramp motor dimensions (this is especially true at the edge output colors). In the poweramp, the increased dimensionality makes it too expensive to do a full multidimensional tuning procedure. Instead we employ an **iterative procedure** as diagrammed below.



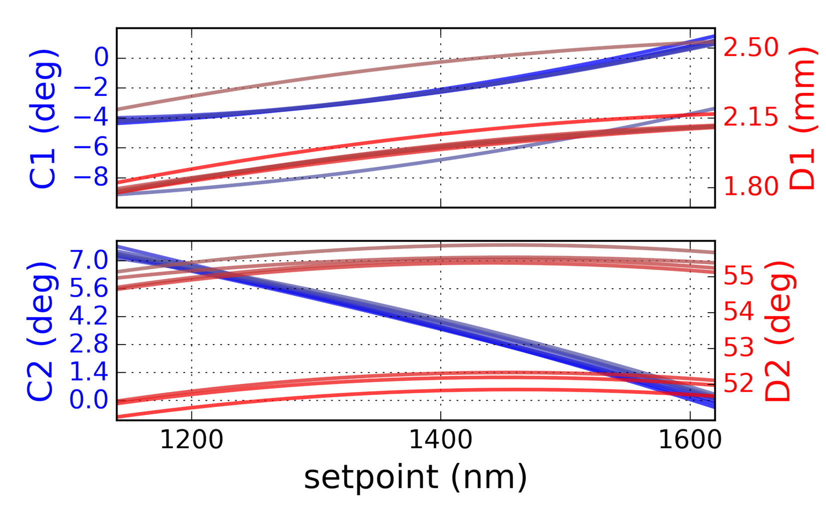

We always end the iteration(s) with C2 so that the OPA's color calibration is best. Typically only one iteration is required but multiple iterations may be necessary if dramatic OPA realignment has occured. In total, poweramp tuning typically takes **less than 1 hour**. Representative procedure output images for D2 (left) and C2 (right) are shown below.




For the D2 figure, the lower panel shows the intensity of the data taken. Note the thick grey line, which represents the chosen points before the final spline step. The top panel compares the old tuning curve (thin) with the output tuning curve (thick). For the C2 image, the bottom panel represents the color of each fit mapped onto detuning. Each separate marker color represents a different setpoint. As with D2, the C2 upper panel compares the old tuning curve (thin black) with the output tuning curve (colored X's).

## Reproducibility

The right-hand plot shows all of the curves generated by this algorithm over the last 4 months. For each motor, increasing color saturation indicates more recent curves. Each new curve corresponds to a new laser alignment. Without calibration, these changes would lead to major artifact in our experiments.




## References

1. Czech, K. J., Thompson, B. J., Kain, S., Ding, Q., Shearer, M. J., Hamers, R. J., … Wright, J. C. (2015). Measurement of Ultrafast Excitonic Dynamics of Few-Layer $MoS_2$ Using State-Selective Coherent Multidimensional Spectroscopy. ACS Nano
2. Boyle, E. S., Neff-Mallon, N. A., & Wright, J. C. (2013). Triply resonant sum frequency spectroscopy: Combining advantages of resonance Raman and 2D-IR. JPC-A