Ionization matrix element retrieval & MF reconstruction from alignment traces

N_2 Results Summary

22nd Oct. 2016

Summary document (Oct. 2017), for full details see:
Bootstrapping to the Molecular Frame with Time-domain Photoionization Interferometry
Marceau et. al. (2017)

Data and documents: https://doi.org/10.6084/m9.figshare.4480349
arXiv: https://arxiv.org/abs/1701.08432

Original experiments, data & data analysis: Claude Marceau

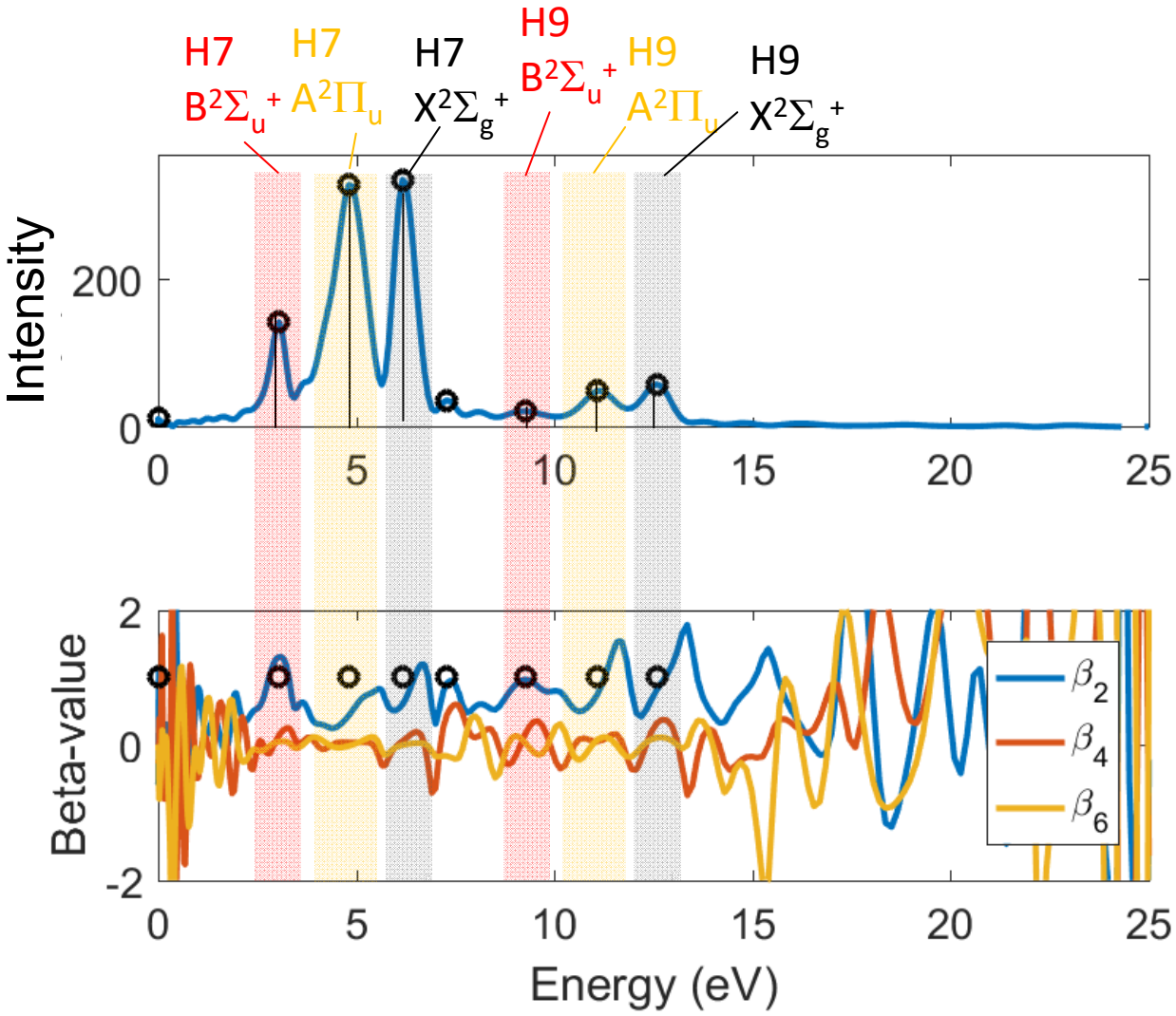
Alignment trace calculation & fitting: Varun Makhija

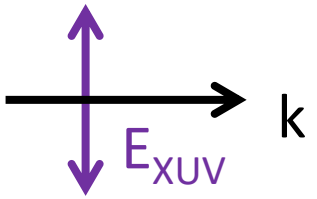
Photoionization calculation & fitting: Paul Hockett

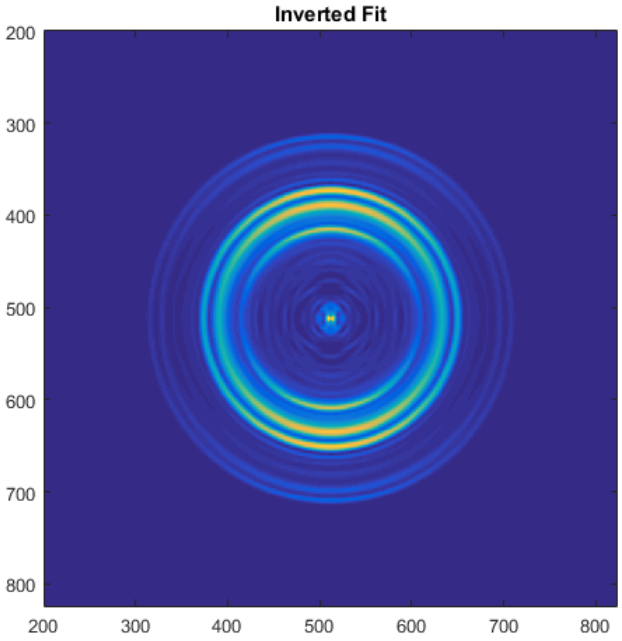
Aims
- VMI with XUV
- Molecular alignment
- Determining the alignment
- Extracting the dipole matrix elements
- Reconstructing the molecular frame

N2 data & spectroscopy

N_2 - Abel inversion results







Varun's fitting (determining the alignment)

Notes from Varun's thesis
Laser-induced rotational dynamics as a route to molecular frame measurements
http://krex.k-state.edu/dspace/handle/2097/18522

where χ becomes the azimuthal angle of the laser polarization vector. Since the Wigner functions $D^j_{m,k}(\theta,\phi,\chi)$ are an irreducible representation on the rotation group SO(3)[52] the angle dependent ion yield can be expanded as

$$S(\theta,\chi) = \sum_{j,k} C_{j,k} D^j_{0,k}(\theta,\chi), \tag{5.1}$$

$$S(\theta,\chi) = \frac{dW}{d\hat{\mathbf{\Omega}}} = \sum_{j,k} C_{j,k} D^j_{0,k}(\theta,\chi),$$
$$C_{j,k} = \sum_{l,m,\lambda,\lambda'} \langle l,-m;l,m|j,0\rangle \langle l,-\lambda;l,\lambda'|j,k\rangle A_{l,\lambda} A_{l,\lambda'} d_{l,m}, \tag{5.5}$$

$$S(t) = \int \rho(\theta,\chi,t) S(\theta,\chi) \sin\theta d\theta d\chi = \sum_{jk} C_{j,k} \int \rho(\theta,\chi,t) D^j_{0k} \sin\theta d\theta d\chi = \sum_{jk} C_{j,k} \left\langle D^j_{0k} \right\rangle (t). \tag{5.7}$$

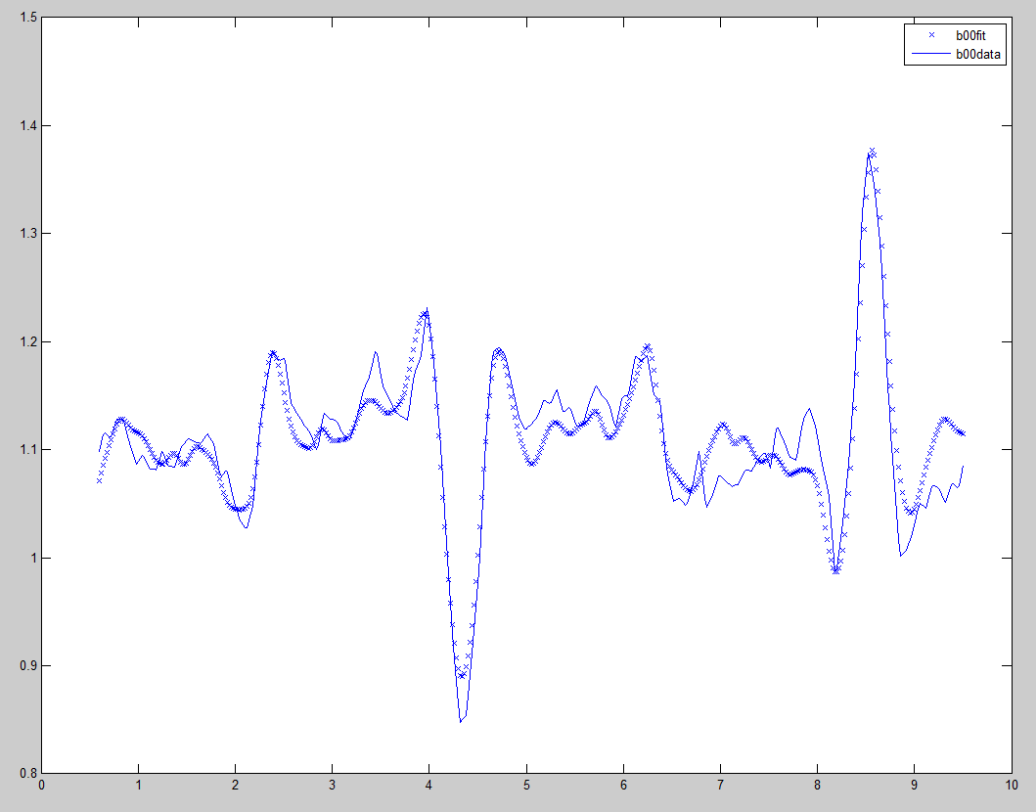
Measured time-dependent signal

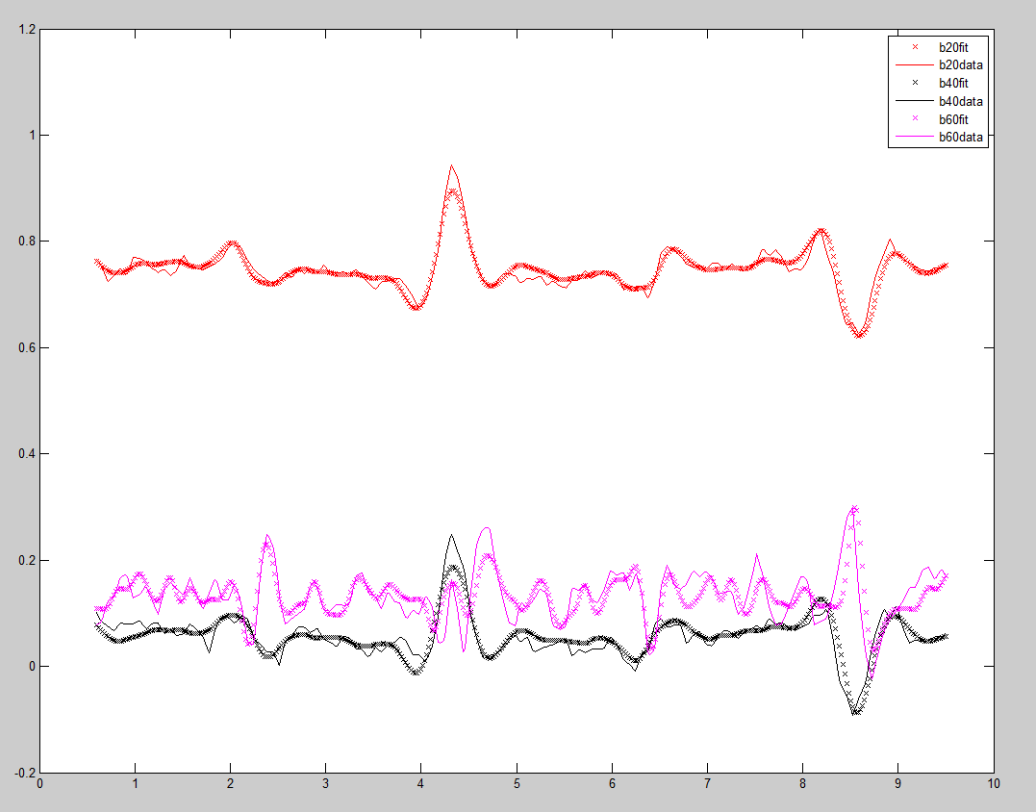
Fitted

Calculated from rotational wavepacket simulations
Function of laser parameters & rotational temperature

Varun’s fitting (determining the alignment)

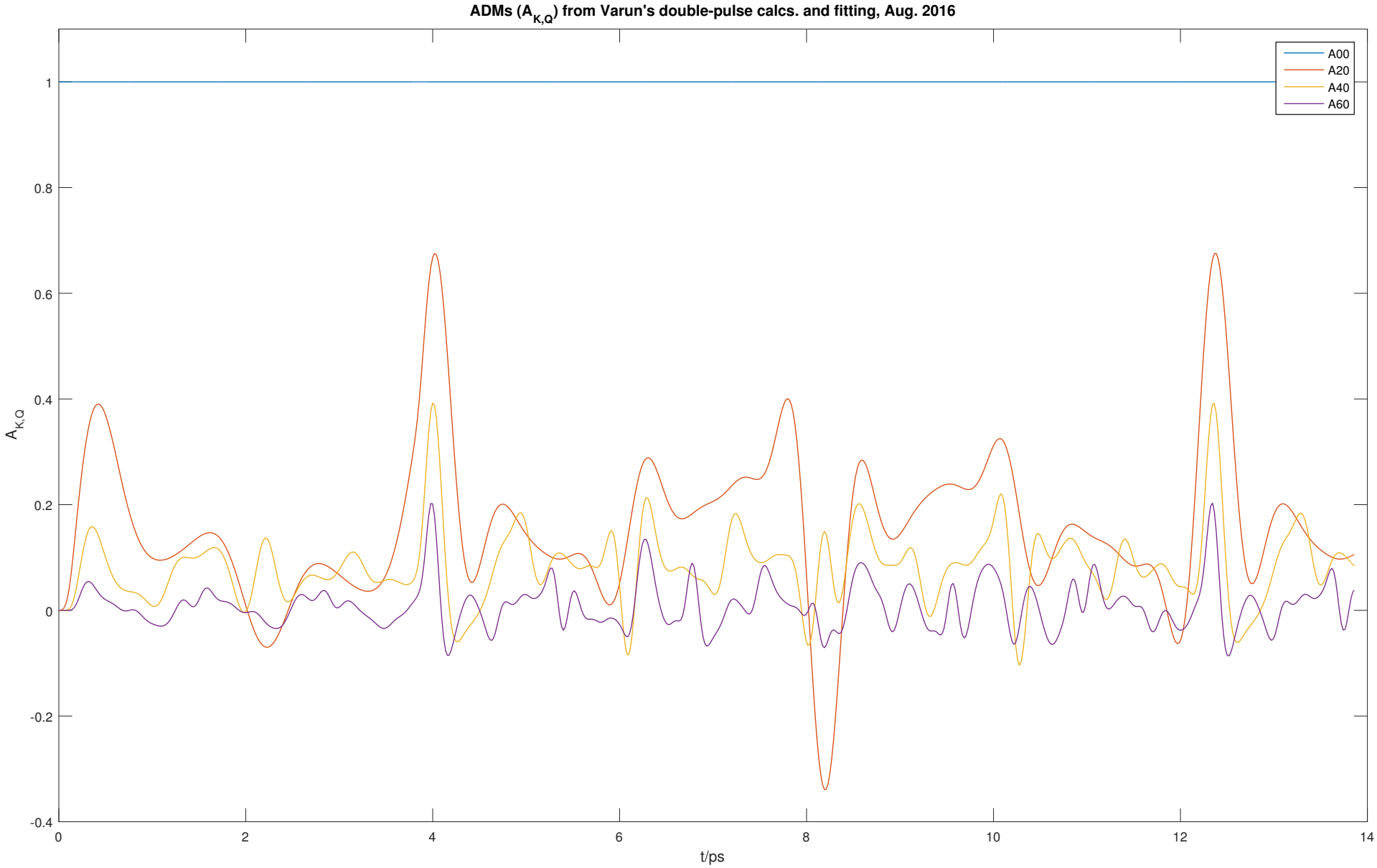
Data - double-pulse alignment data, X-state H5





Varun’s fitting (determining the alignment)

The alignment is characterised by axis distribution moments (ADMs) defining the alignment in terms of spherical harmonic expansion coefficients.



Paul's fitting (determining the photoionization dynamics)

Use known ADMs to determine the unknown dipole matrix elements. Everything else is angular momentum coupling and symmetry parameters; complicated, but analytical.

ADMs from Varun's fit

$$\beta_{L,M}(t) = (2L+1)^{1/2} \sum_P (-1)^P \begin{pmatrix} 1 & 1 & P \\ p & -p & R \end{pmatrix} e_{-p} e^*_{-p}$$
$$\times \sum_K \sum_Q (2K+1)^{1/2} \begin{pmatrix} P & K & L \\ Q-M & -Q & M \end{pmatrix} A_{K,-Q}(t)$$
$$\times \sum_{q,q'} (-1)_{q'} \begin{pmatrix} 1 & 1 & P \\ q & -q' & q'-q \end{pmatrix} \begin{pmatrix} P & K & L \\ q-q' & q'-q & 0 \end{pmatrix}$$
$$\times \sum_{l,l'} \sum_{\lambda,\lambda'} (-1)^{\lambda'} (2l+1)^{1/2} (2l'+1)^{1/2} \begin{pmatrix} l & l' & L \\ \lambda & -\lambda' & M \end{pmatrix} \begin{pmatrix} l & l' & L \\ 0 & 0 & 0 \end{pmatrix}$$
$$\times (-i)^{l'-l} \sum_{\Gamma,\Gamma'} \sum_{\mu,\mu'} \sum_{h,h'} b^{\Gamma\mu*}_{hl\lambda} b^{\Gamma'\mu'}_{h'l'\lambda'} \boldsymbol{D}^{\Gamma\mu*}_{hl}(q) \boldsymbol{D}^{\Gamma'\mu'}_{h'l'}(q'). \quad (1$$

Measured angular parameters

Matrix elements to determine

See *General phenomenology of ionization from aligned molecular ensembles* (Hockett, NJP, 17 023069 2015) for details.

Paul's fitting (determining the photoionization dynamics)

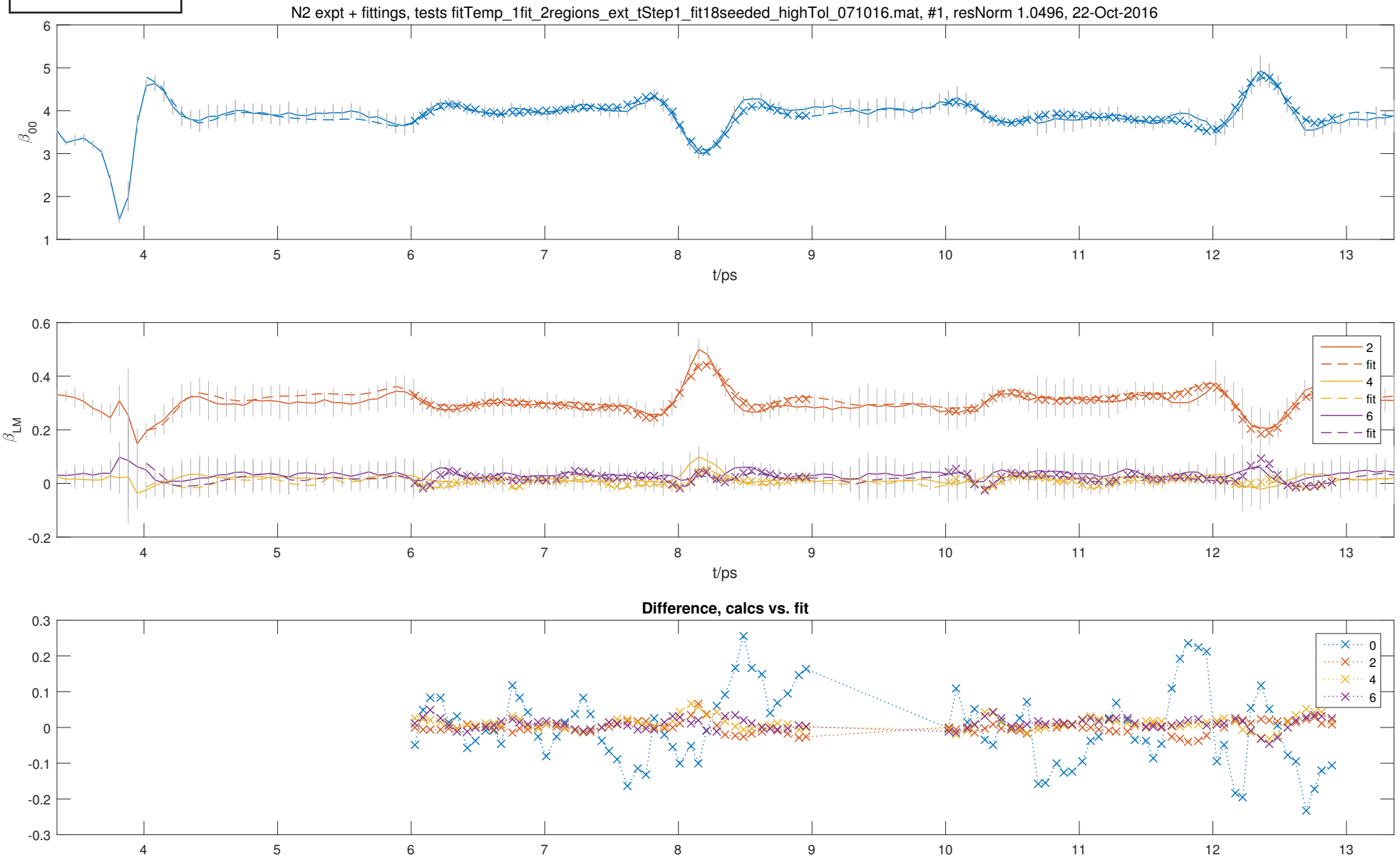
Inputs:
Data - Claude's updated analysis August 2016
Alignment calculations - from Varun's analysis, 29th August 2016, as per above details.

Basic method:

- Renormalize beta values (from Legendre polynomial to spherical harmonic expansion)
- Fitting to determine symmetrized matrix elements
- Statistical sampling of fitting hyperspace via multiple coarse fits with random start vectors (approx. 1 hour per fit)
- Bootstrapping fits via gradually adding additional data points and/or tightening tolerances (multiple hours per fit)

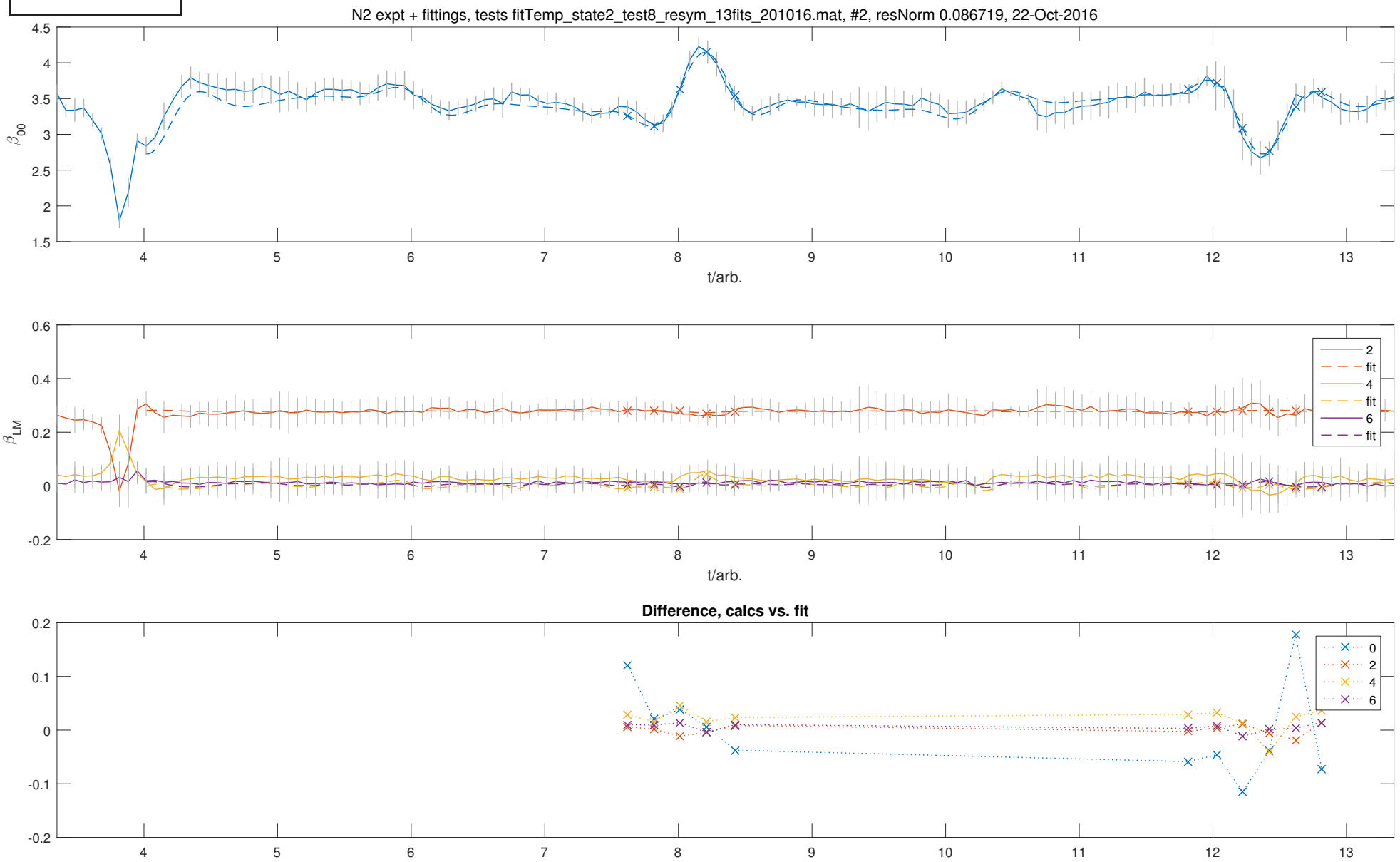
X-state, H5

Example results, ‘x’ marks points included in the fitting
Experimental uncertainties may be incorrectly renormalised here!



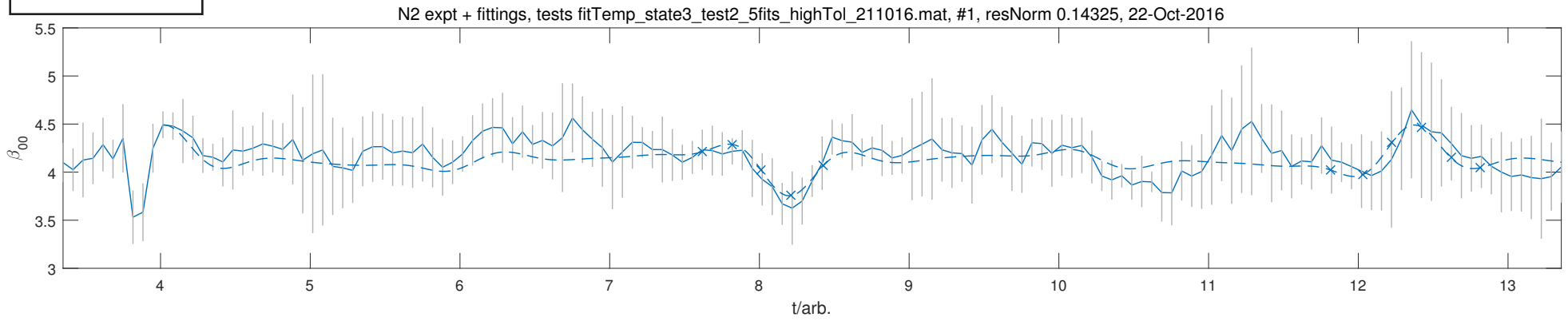
A-state, H5

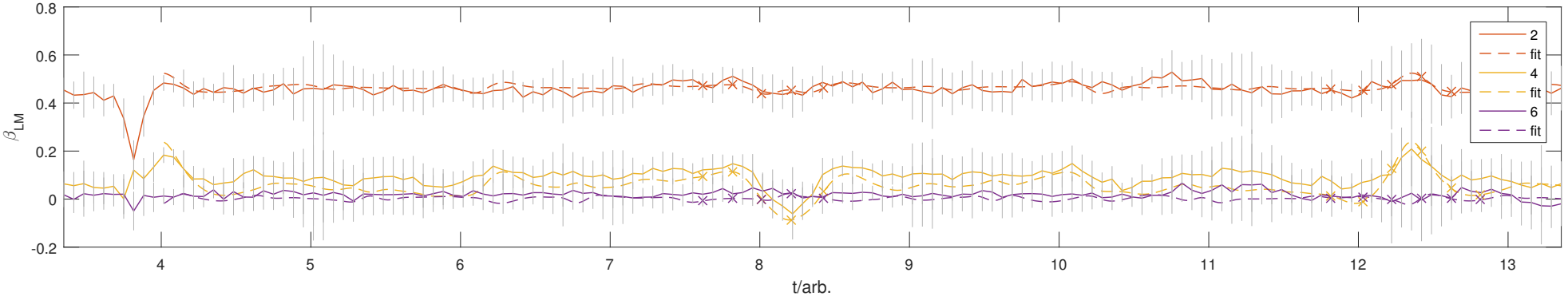
Example results, ‘x’ marks points included in the fitting
Experimental uncertainties may be incorrectly renormalised here!

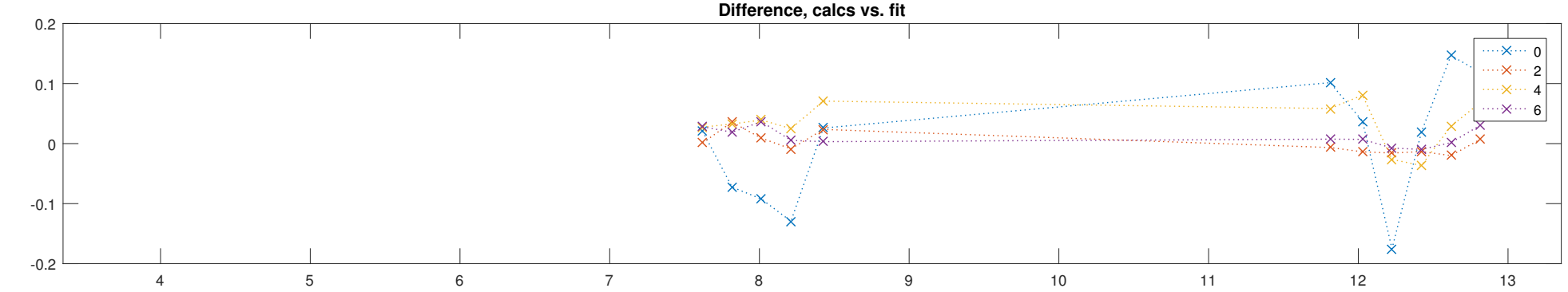


B-state, H5

Example results, ‘x’ marks points included in the fitting
Experimental uncertainties may be incorrectly renormalised here!



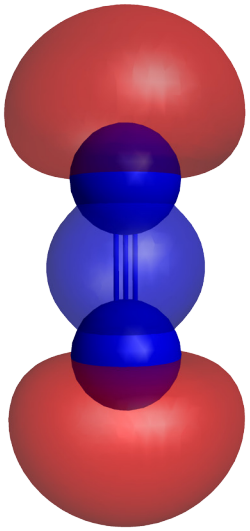




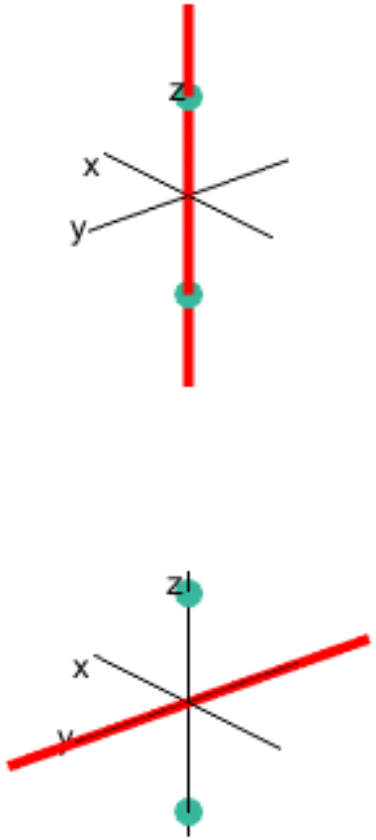
Molecular frame reconstruction

From the matrix elements determined the full MFPAD can be obtained...
(via another big eqn. similar to the above)

X-state, H5

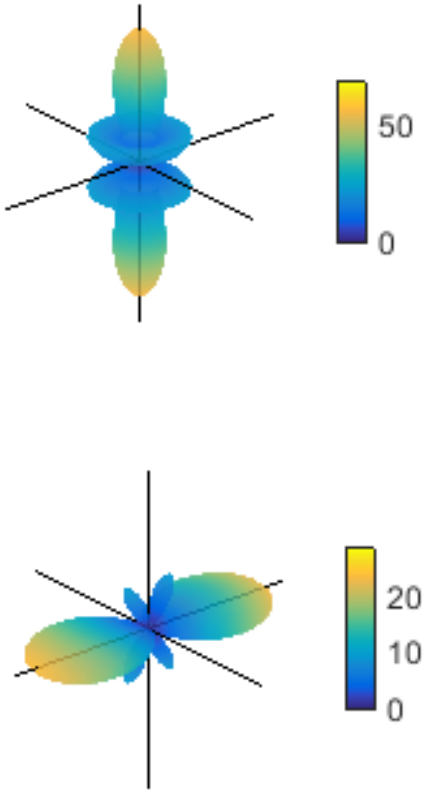


Ionizing orbital
$3\sigma_g$



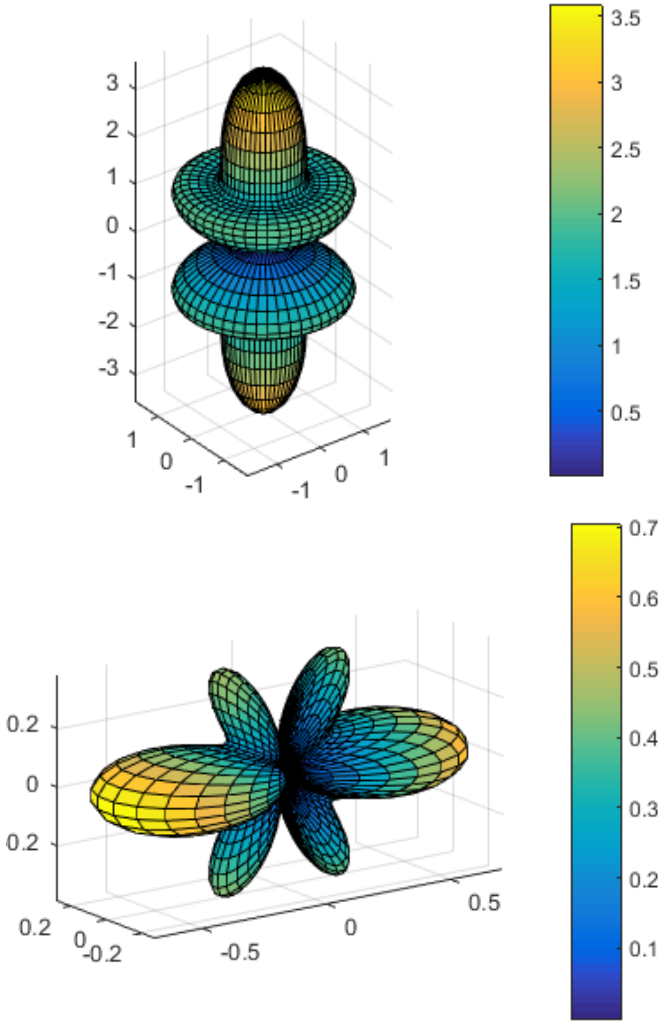
Mol. frame
polarization geometry

8.50 eV



ePS result
(note energy slightly different)

7.7 eV



Determined from fit result

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