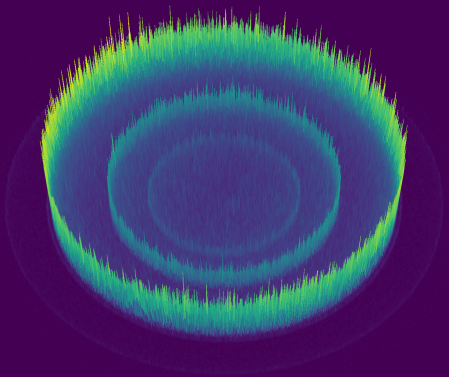
Photodetachment of O^- yielding $O(^1D_2, ^3P)$ atoms, viewed with velocity-map imaging

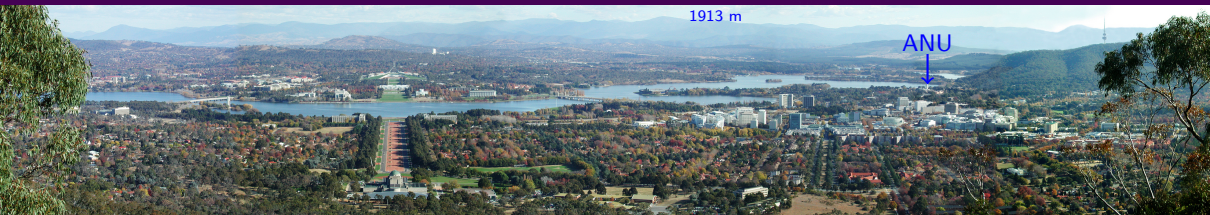
Steve Gibson, Ben Laws, and Brenton Lewis

LPC/RSPE Australian National University, Canberra

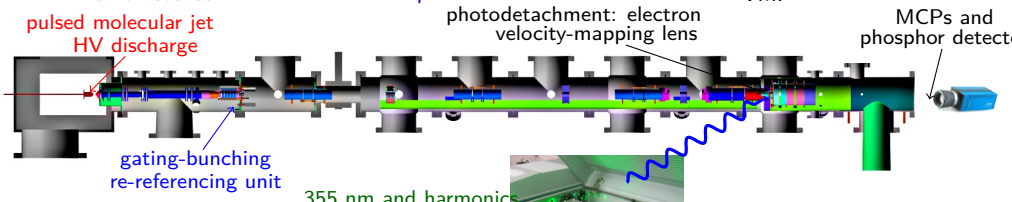
ISMS June 23, 2016

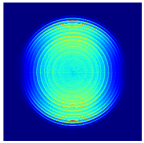


- Photodetachment/VMI
- PES/PAD $O^- \rightarrow O(^3P_J)$
- R-Matrix calculation
- PES/PAD $O^- \rightarrow O(^1D_2)$



Spectrometer - photodetachment/photofragmentation





355 nm and harmonics
Nd:YAG laser

OPO laser 220 nm−1750 nm

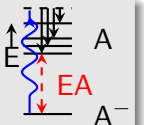
Fast beam spectrometer:
Cyr PhD Thesis (UC Berkeley 1993)

Velocity-map imaging lens:
Eppink and Parker Rev Sci Instrum **68** 3477 (1997)

Gating-bunching-rereferencing unit:
(ANU) Dedman *et al.* Rev Sci Instrum **73** 2915 (2001)

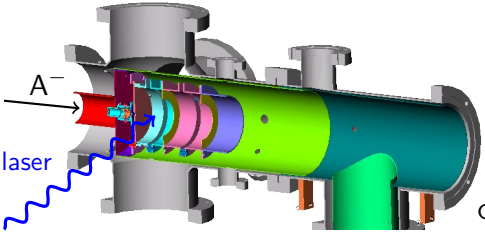
Photodetachment:

$A^- + h\nu \rightarrow A^* + e^-$

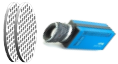


Velocity-map imaging

VMI lens coaxial with ion-beam

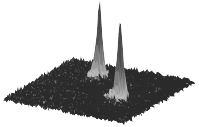


MCPs+phosphor

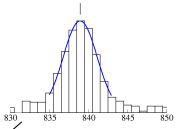


CCD camera 2048x2048 pixel

monochrome camera image



(x_i, y_i)



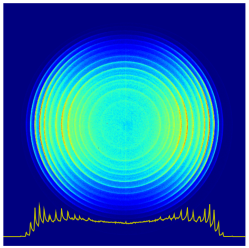
sub-pixel centroid

bin to image

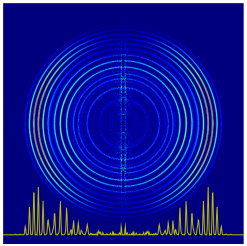
circularize Gascooke (Flind U)

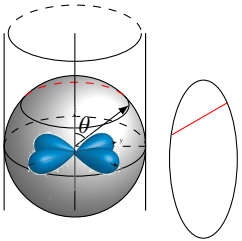
e^- trajectories

2D projection O_2^-



3D slice





$\frac{\Delta E}{E} \sim$ constant $\rightarrow e^-$ speed

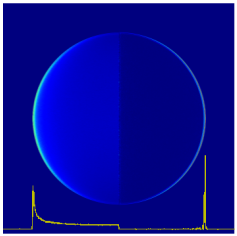
Inverse Abel transformation: **PyAbel**: https://github.com/PyAbel

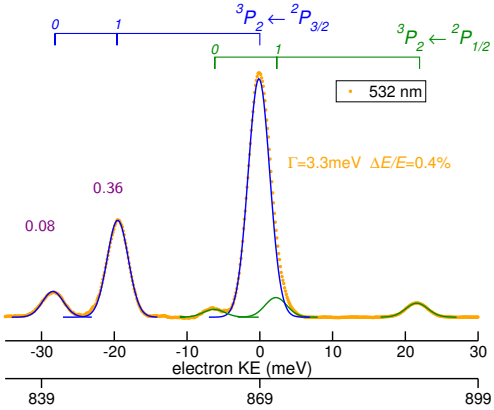
O^- photodetachment

Photoelectron spectrum (PES)

$$O^-(^2P_{J''}) + h\nu(532\text{ nm}) \rightarrow O(^3P_{J'}) + e^- \ (869\text{ meV})$$

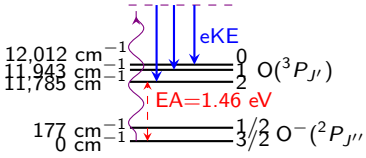
VMI 3D slice



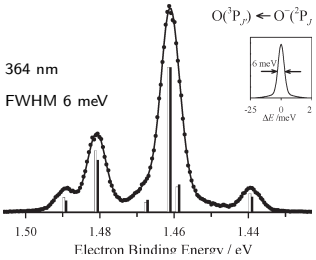


(ANU) Cavanagh *et al.* PRA **76** 052708 (2007)

Spectroscopy	
position (cm^{-1}):	227, 158, 0, (-177)
width (meV):	3.3
intensity ratio:	0.08, 0.36
temperature (K):	200



Ervin *et al.* J Phys Chem A**107** 8521 (2003)



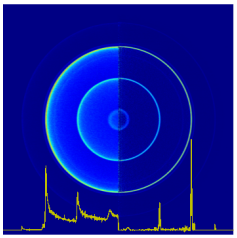
$$eBE = h\nu - eKE$$

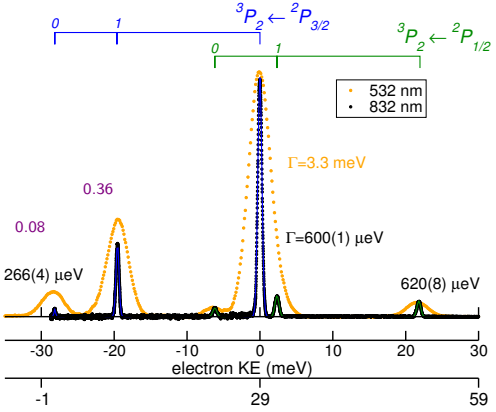
$1\text{ meV} \sim 8\text{ cm}^{-1}$

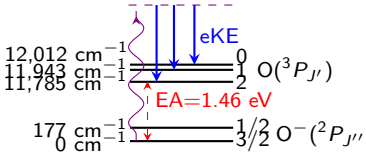
O^- photodetachment

Photoelectron spectrum (PES)

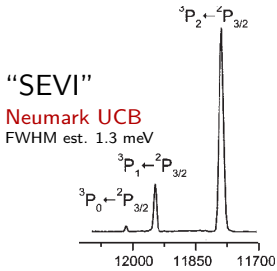
$$O^-(^2P_{J''}) + h\nu(532{,}832\text{ nm}) \rightarrow O(^3P_{J'}) + e^- \ (869{,}29\text{ meV})$$



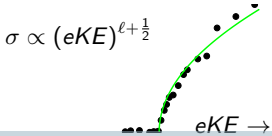




Garand *et al.* J Phys Chem A**113** 4631 (200



Wigner Phys Rev **73** 1002 (1948)



(ANU) Cavanagh *et al.* PRA **76** 052708 (2007)

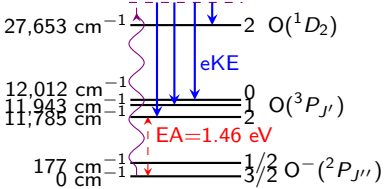
Spectroscopy		
position (cm^{-1}):	227, 158, 0, (-177)	227, 158, 0, (-177)
width (meV):	3.3	0.3−0.6
intensity ratio:	0.08, 0.36	0.01, 0.2!
temperature (K):	200	211

$$\sigma^{TI}(J_a J_i) = \frac{4\pi^2\omega}{3c} \sum_{\ell_c} \begin{pmatrix} \ell_c & 1 & \ell_i \\ 0 & 0 & 0 \end{pmatrix}^2 (\epsilon\ell_c|r|n_i\ell_i)^2 [J_a, S_i, L_i] \times$$

$$N(S_a L_a, \ell_i|\} S_i L_i)^2 \sum_{j_i} [j_i] \begin{Bmatrix} S_a & L_a & J_a \\ s & \ell_i & j_i \\ S_i & L_i & J_i \end{Bmatrix}^2 ,$$

For oxygen atom:

$$^3P_{2,1,0}: \quad L_a = 1 \quad S_a = 1 \quad J_a = 2, 1, 0$$
$$^2P_{\frac{1}{2},\frac{3}{2}}: \quad L_i = 1 \quad S_i = \frac{1}{2} \quad J_i = \frac{1}{2}, \frac{3}{2}$$



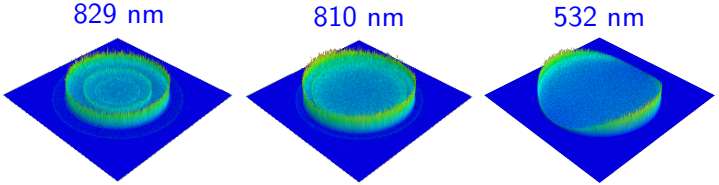
and the $9j$ symbol sum gives:

$$^3P_2 \leftarrow {}^2P_{\frac{3}{2}} : {}^3P_1 : {}^3P_0 = 1 : 0.36 : 0.08$$

^3P ratio $\quad {}^2P_{\frac{1}{2}} : {}^2P_{\frac{3}{2}} = 0.4$

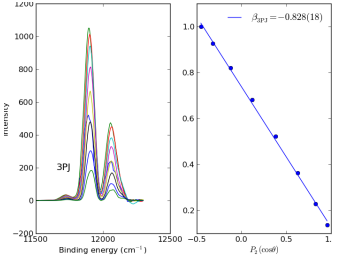
$$\text{Temperature } (O^-) = \frac{-177.13 \times hc}{k \ln(5 \times \frac{I_{\frac{1}{2}}}{I_{\frac{3}{2}}})}$$

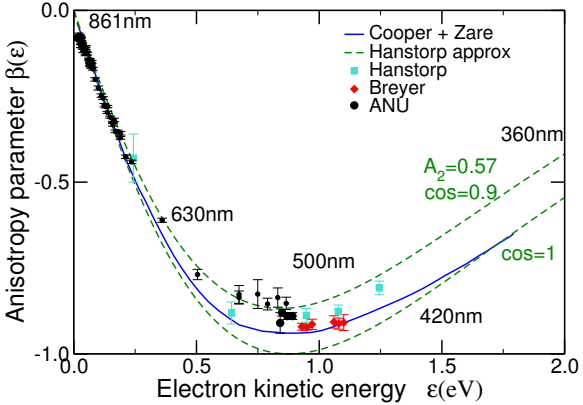
O^- photoelectron angular distribution (PAD)



$$I(\theta,\epsilon) = \frac{\sigma(\epsilon)}{4\pi}[1+\beta(\epsilon)P_2(\cos\theta)]$$

β anisotropy parameter





p-orbital electron, $\Delta\ell = \pm 1$

$\sigma \propto R_s^2 + 2R_d^2$ (incoherent sum)

$\beta \propto \dfrac{2\left(R_d^2 - 2R_dR_s\right)}{\sigma}$ (coherent sum)

Hanstorp approx: $A_2\epsilon \sim R_d/R_s$

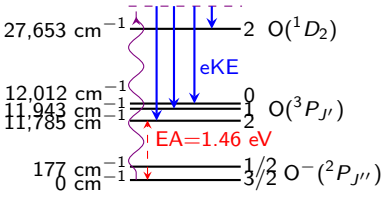
Δ = partial wave phase shift

$$\beta_{l=1}(\epsilon) = \frac{2(A_2\epsilon)^2 - 4(A_2\epsilon)\cos(\Delta)}{1+2(A_2\epsilon)^2}$$

PAD for photodetachment of O^- yielding $O(^1D_2)$ atoms?

A "special" case?

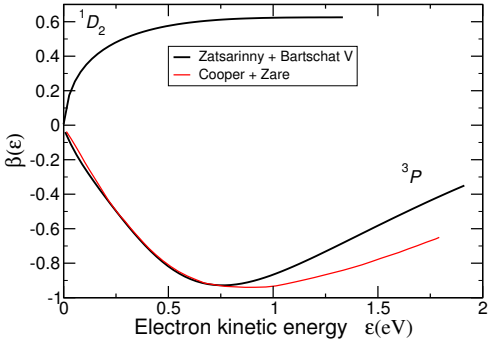
$$O^-(^2P_{\frac{3}{2},\frac{1}{2}}) + h\nu \rightarrow O(^3P_{2,1,0}, {}^1D_2) + e^-$$

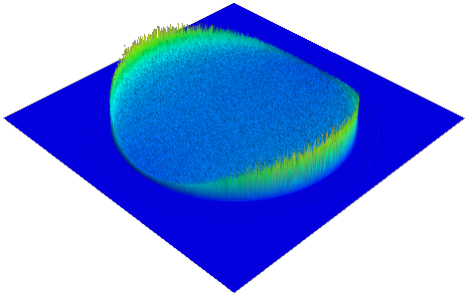


B-spline R-matrix calculation

Oleg Zatsarinny and Klaus Bartschat, Drake U. Iowa USA

$^1D_2$ prediction





R-matrix code: Burke and Berrington

"... have reached a level of stability and robustness ... used to solve a wide variety of low electron and positron scattering problems" Morgan *et al* Comp Phys Comm **114** 120-128 (1998)

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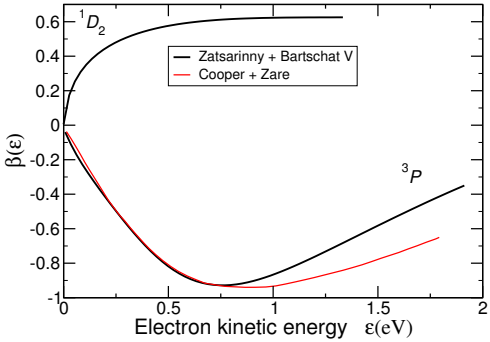

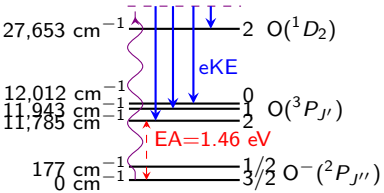

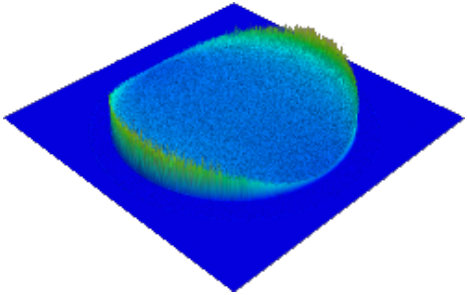

R-matrix code: Burke and Berrington

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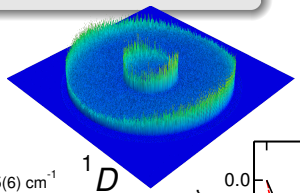
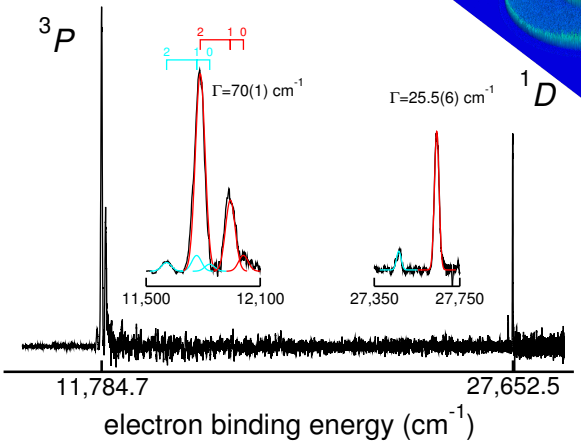
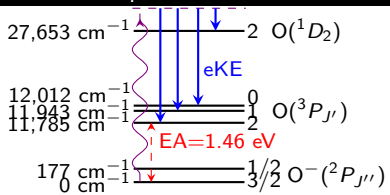
Photodetachment of O^- yielding $O(^1D_2)$ atoms

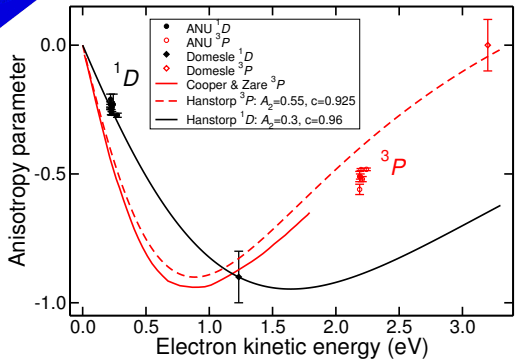
"same but different"

$$O^-(^2P_{\frac{3}{2},\frac{1}{2}}) + h\nu(340\text{ nm}) \rightarrow O(^3P_{2,1,0}, {}^1D_2) + e^-(2.2,0.2\text{ eV})$$



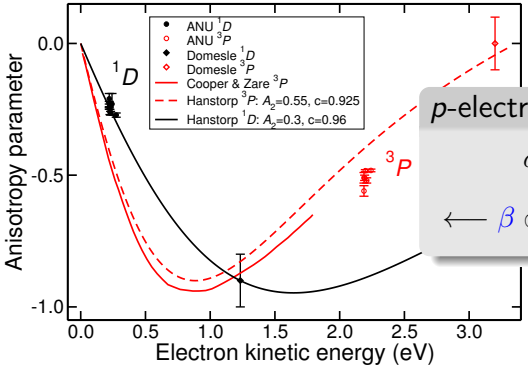






$\beta_{^1D_2} < 0$ maps a different isotropy curve, slower onset, higher-energy minimum than $\beta_{^3P}$

Calculations R-matrix



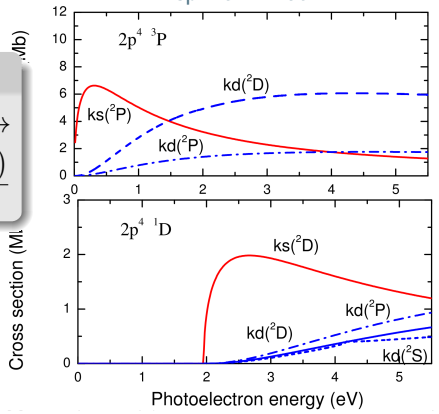
p-electron $\Delta\ell = \pm 1$

$$\sigma \propto R_s^2 + 2R_d^2 \longrightarrow$$

$$\longleftarrow \beta \propto \frac{2\left(R_d^2 - 2R_d R_s\right)}{\sigma}$$

Zatsarinny Phys Rev A **73** 022714 (2006)

B-spline R-matrix



Data consistent with σ calculation, slower onset of d-wave cross section for the O($^1D_2$) channel

Issue with R-matrix code

Oleg (Drake): I still get positive values for β_{1_D}, even after double check of possible bugs in the program
Klaus (Drake): The Belfast R-matrix code . . . had some interesting troubles
Mabbs (Wash U): . . . to get a sensible low eKE curve we have to change a sign in the calculation

Back to basics: Zero-core-contribution model

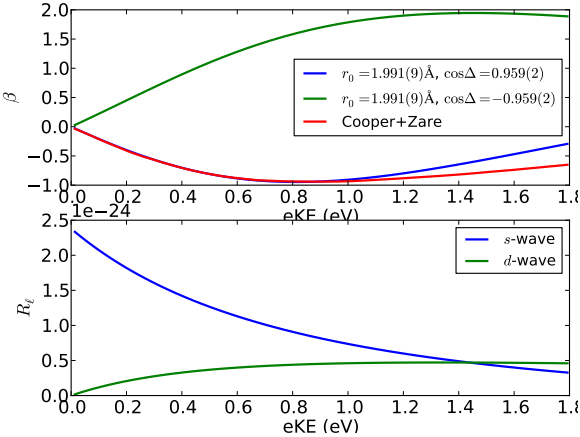
Stehman and Woo Phys Rev A20 281-290 (1979)

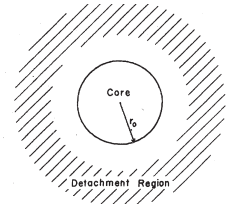
- Evaluate R_s and R_d matrix elements using H-atom ground-state wavefunction for initial state and plane-wave for free electron
- Simple analytical expressions for R_s and R_d
- Two parameter model: r_0 and $\gamma = \cos(\Delta)$

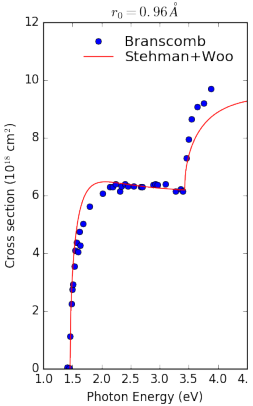
$$\sigma \propto R_s^2 + 2R_d^2$$

$$\beta \propto \frac{2(R_d^2 - \gamma R_d R_s)}{\sigma}$$

$$\frac{d\sigma}{d\Omega} \propto |\langle k|\hat{e}\cdot\mathbf{r}|0\rangle|^2$$

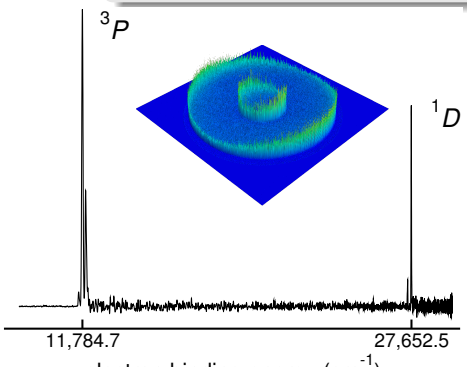


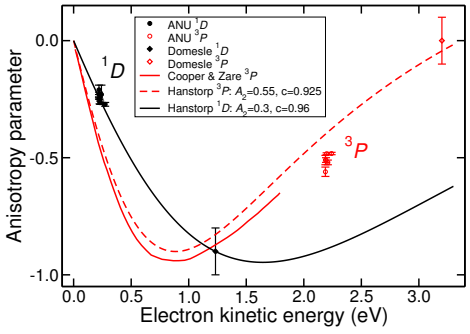




Conclusions

- VMI provides μeV electron kinetic-energy resolution and full angular distribution
- $R-$matrix computational code predicts $\beta_{^1D_2} > 0$
- Experiment
  - $\beta_{^1D_2}$ same sign as $\beta_{^3P}$
  - $\beta_{^1D_2}$ different energy dependence to $\beta_{^3P}$
- Measurements consistent with "simple" s- and $d-$wave detachment model
- "Black box" R-matrix computational code may have problems ...





Acknowledgements

Other people

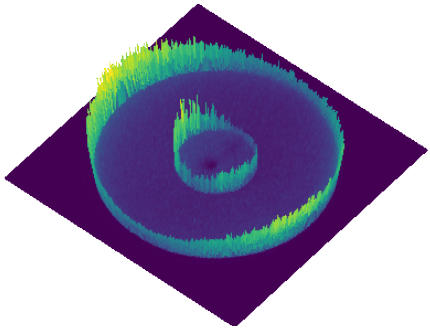
Ly Duong
Hons. 2012

Steve Cavanagh

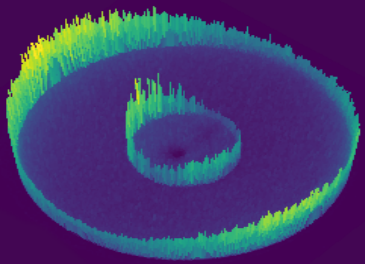
Brenton Lewis

and technical assistance: *Colin Dedman*, *Kevin Lonsdale*, *Ros Tranter*, *Steve Battisson*

Oleg Zatsarinny and Klaus Bartschat, Drake U. Iowa USA



End



Appendix

- PyAbel: Abel transforms
- Cooper-Zare angular distribution of photoelectrons

PyAbel: Abel transform software
https://github.com/PyAbel

https://github.com/PyAbel/PyAbel

Search

ting Started | scipy array tip sheet | Fig. 7. The absolution… | git remote | python uninstall | afl | Clo

README.rst

PyAbel README

build error | BUILD PASSING

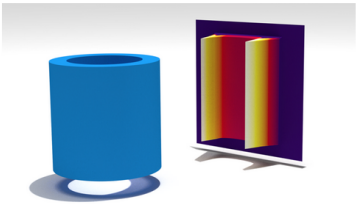
Note: This readme is best viewed as part of the PyAbel Documentation.

Introduction

`PyAbel` is a Python package that provides functions for the forward and inverse Abel transforms. The forward Abel transform takes a slice of a cylindrically symmetric 3D object and provides the 2D projection of that object. The inverse abel transform takes a 2D projection and reconstructs a slice of the cylindrically symmetric 3D distribution.

Inverse Abel transforms play an important role in analyzing the projections of angle-resolved photoelectron/photoion spectra, plasma plumes, flames, and solar occultation.

PyAbel provides efficient implementations of several Abel transform algorithms, as well as related tools for centering image symmetrizing images, and calculating properties such as the radial intensity distribution and the anisotropy parameters.



Transform Methods

The outcome of the numerical Abel Transform depends on the exact method used. So far, PyAbel includes the following transform methods:

1. `basex` - Gaussian basis set expansion of Dribinski and co-workers.
2. `hansenlaw` - recursive method of Hansen and Law.
3. `direct` - numerical integration of the analytical Abel transform equations.
4. `two_point` - the "two point" method of Dasch and co-workers.
5. `three_point` - the "three point" method of Dasch and co-workers.
6. `onion_peeling` - the "onion peeling" deconvolution method of Dasch and co-workers.
7. `onion_bordas` - "onion peeling" or "back projection" method of Bordas *et al.* based on the MatLab code by Rallis and Wells *et al.*
8. `linbasex` - the 1D-spherical basis set expansion of Gerber *et al.*
9. `fh` - Fourier–Hankel method (not yet implemented).
10. `pop` - polar onion peeling method (not yet implemented).

Cooper and Zare J Chem Phys **48** 942 (1968)

$$\beta_\ell = \frac{\ell(\ell-1)R_{\ell-1}^2 + (\ell+1)(\ell+2)R_{\ell+1}^2 - 6\ell(\ell+1)R_{\ell+1}R_{\ell-1}\cos(\delta_{\ell+1}-\delta_{\ell-1})}{3(2\ell+1)[\ell R_{\ell-1}^2 + (\ell+1)R_{\ell+1}^2]}$$