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A Bayesian model selection framework to probe the structure of Langmuir-Blodgett monolayers from neutron reflectometry

SciML Seminar — 2019/10/17

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let's break that title down

A *Bayesian model selection* framework to probe the structure of *Langmuir-Blodgett monolayers* from *neutron reflectometry*

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Langmuir-Blodgett monolayers

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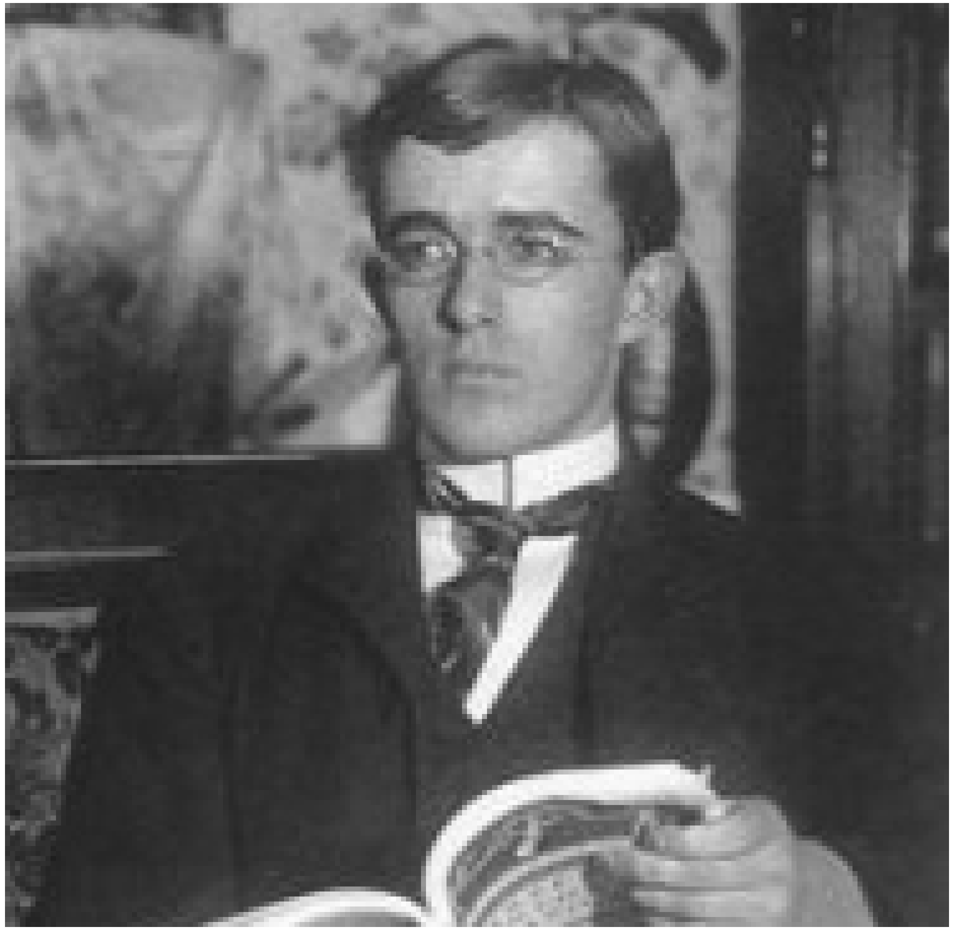
Langmuir/Blodgett



Irving Langmuir

Katharine Blodgett

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amphiphiles

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important biologically and technologically

- lipids: make up cell membranes
- surfactants: present in many consumer products

Liley, *et al.*, Langmuir, **33**, 4301 (2017);
Fisher, *et al.*, in *Neutron Scattering - Applications in Biology, Chemistry, and Materials Science* (Academic Press, 2017) pp. 1-75

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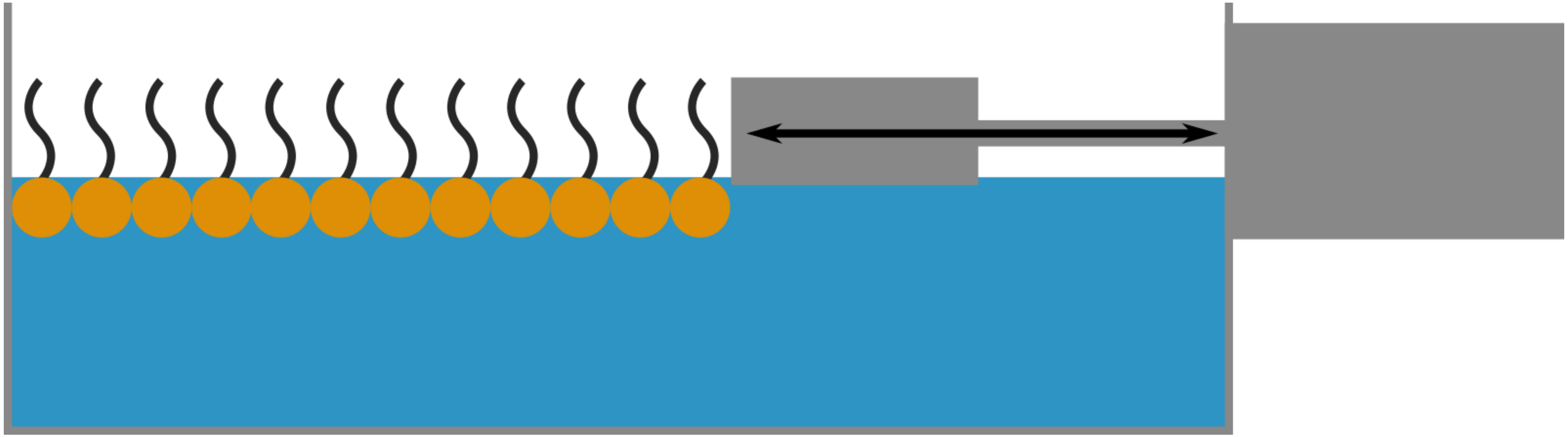
self-assemble



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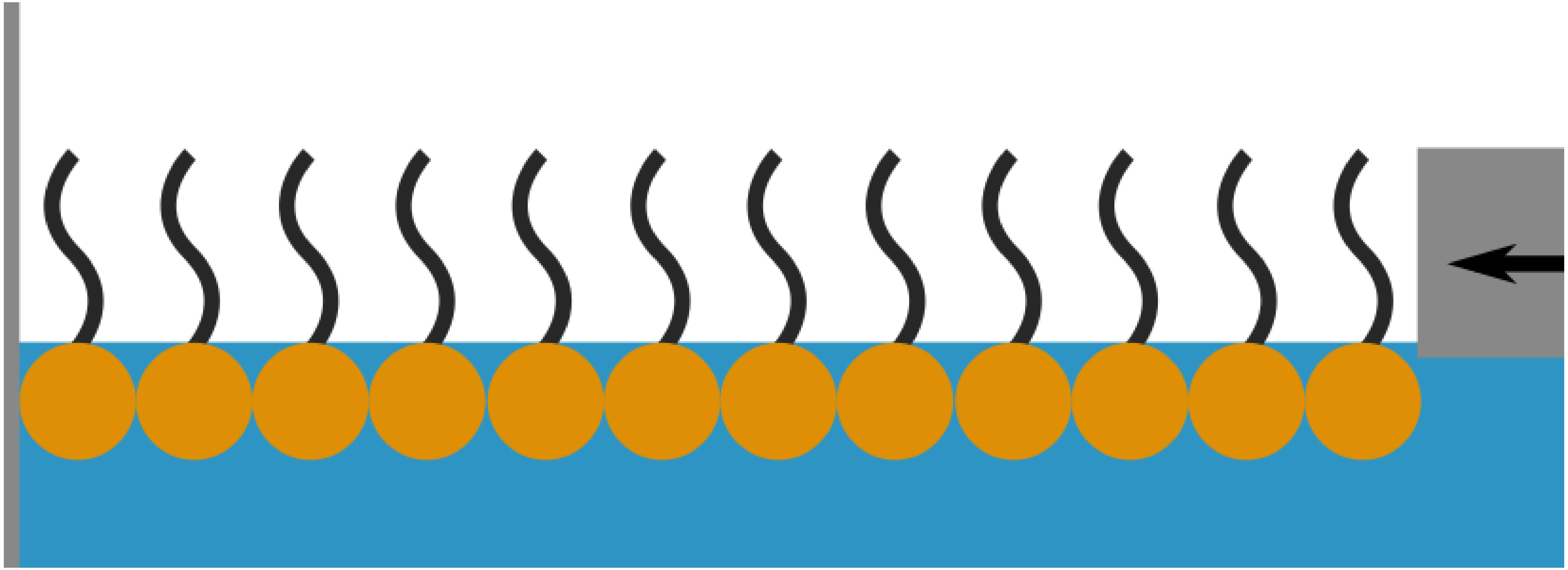
structure is important



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neutron reflectometry

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neutron reflectometry

- Non-invasive measurement of structure at an interface
- Application to Langmuir-Blodgett monolayers only developed in the past thirty years
- Popularised through the work of Penfold, Thomas and others

Grundy, *et al.*, Thin Solid Films, **159**, 43 (1988); Penfold and Thomas, J. Phys. Condens. Matter, **2**, 1369 (1990); Penfold and Thomas, Curr. Opin. Colloid Interface Sci., **19**, 198 (2014)

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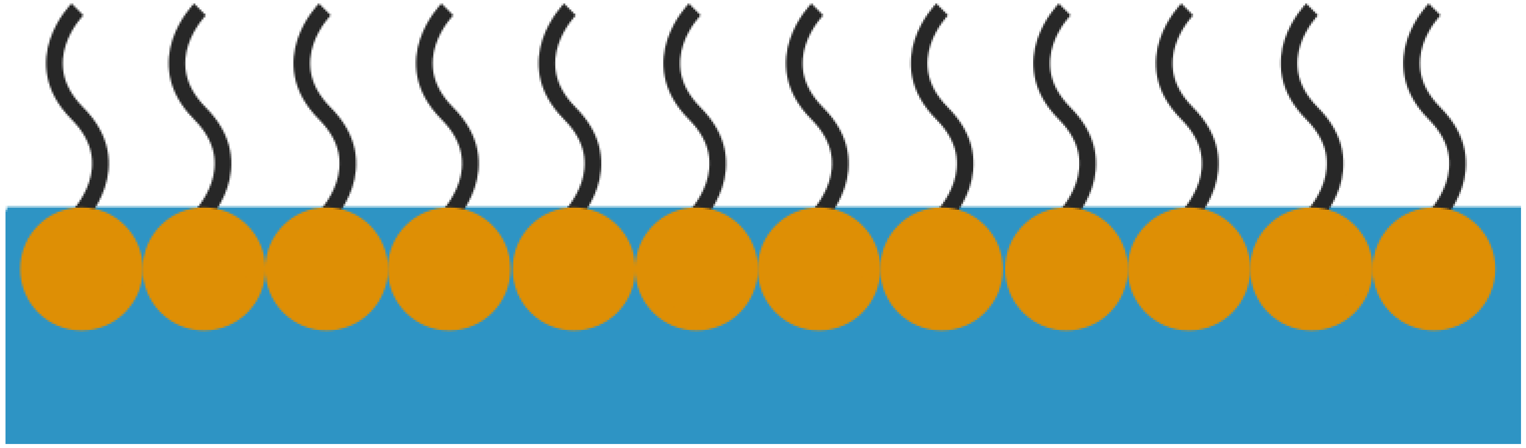

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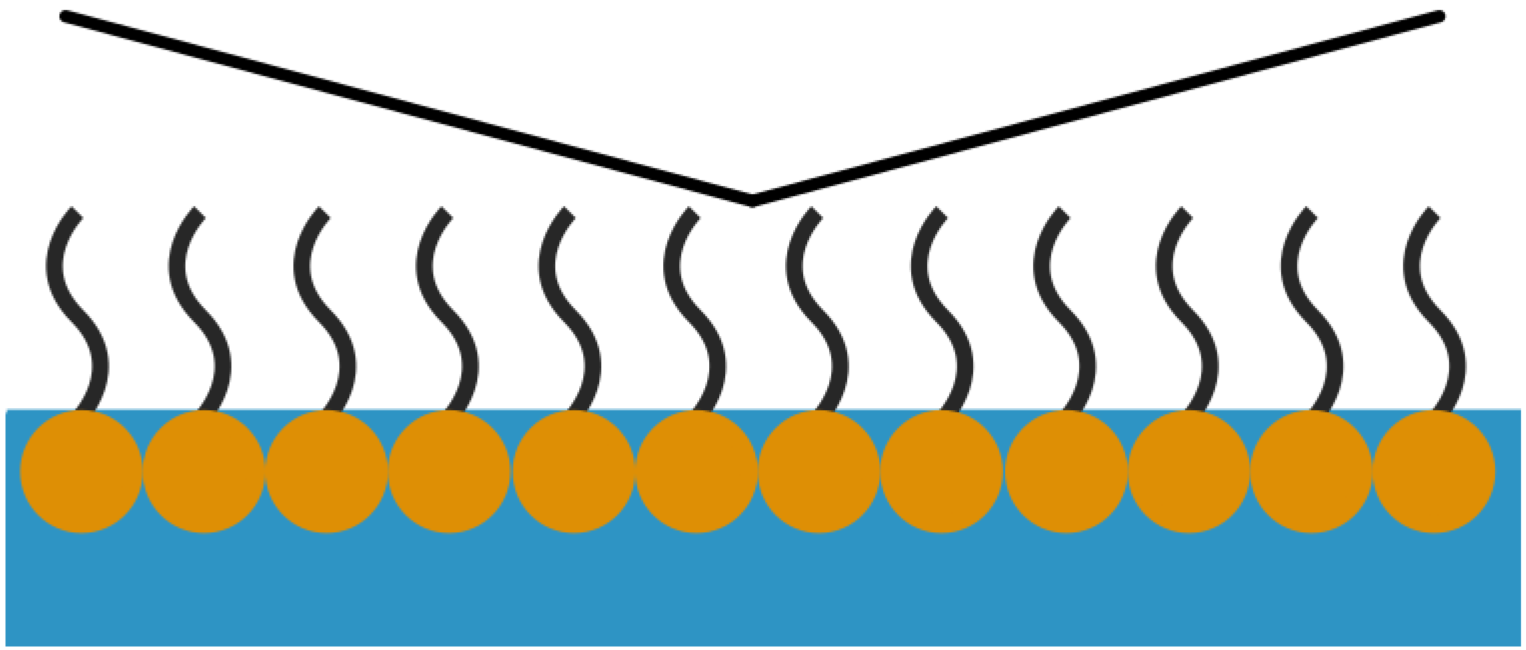

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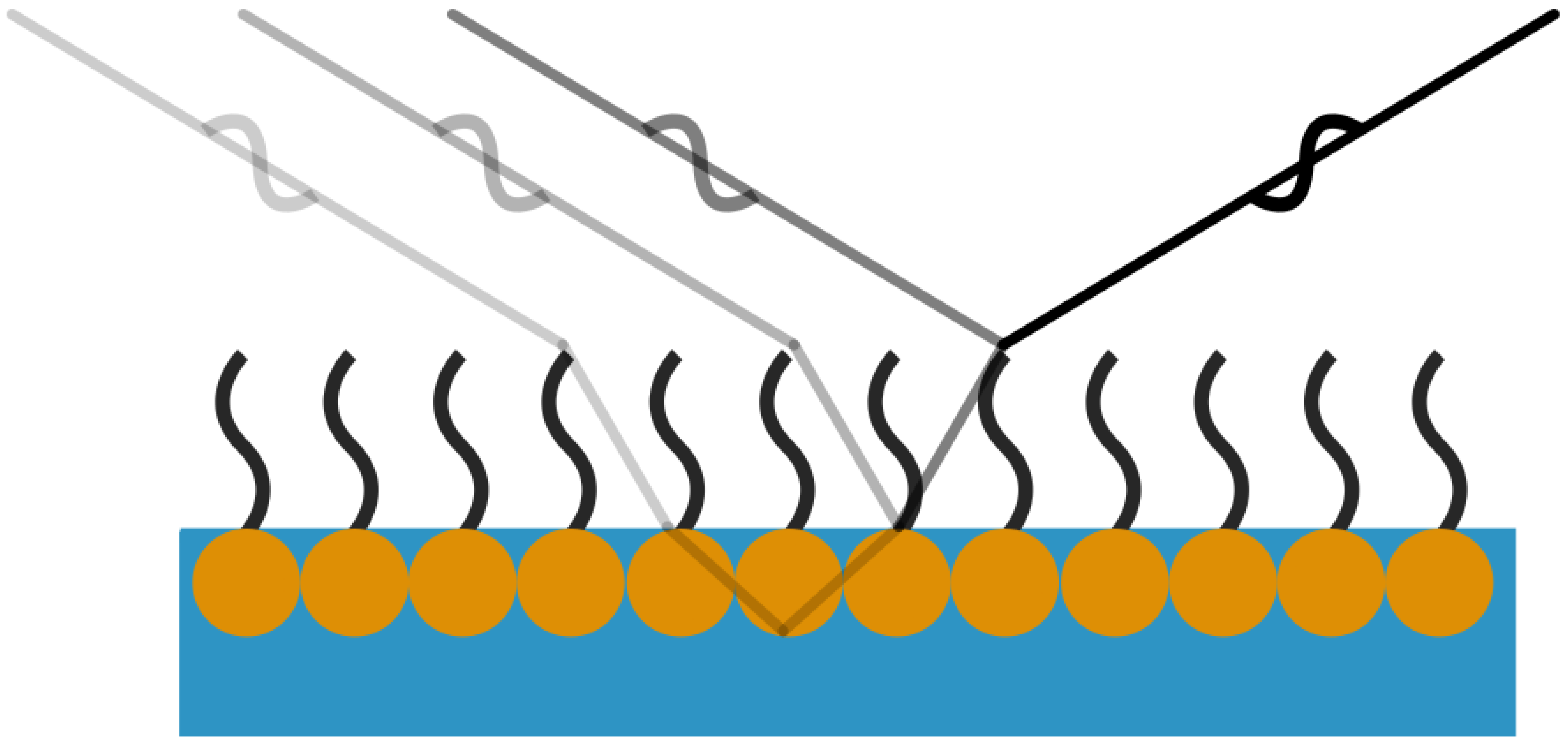

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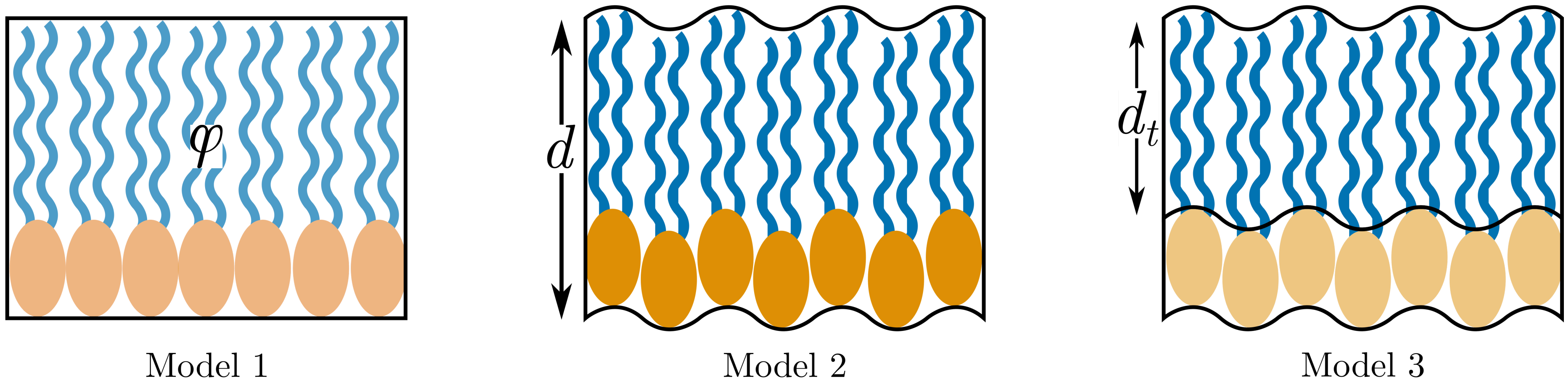

Campbell, *et al.*, J. Colloid Interface Sci., **531**, 98 (2018)

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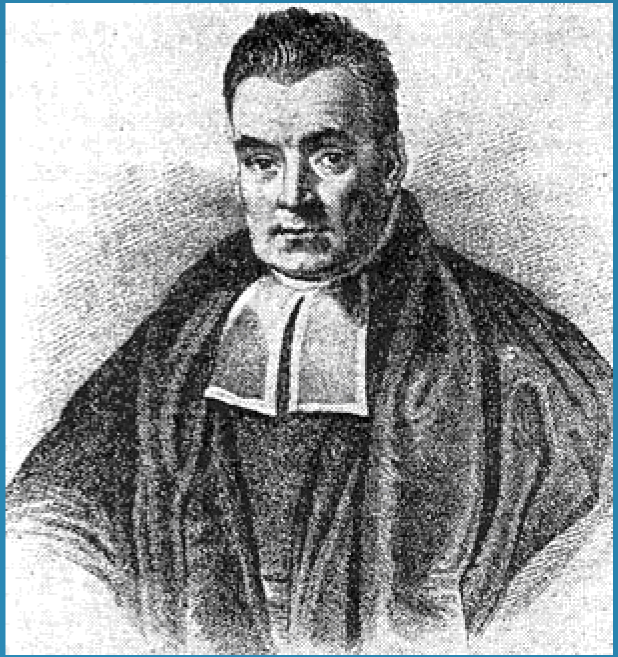
Bayesian model selection



❤️ Thomas Ba(e)yes ❤️
English statistician; 1701-1761

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Bayes theorem

$$p(H|\mathbf{D},\mathbf{I}) = \frac{p(\mathbf{D}|H,\mathbf{I})p(H|\mathbf{I})}{p(\mathbf{D}|\mathbf{I})}$$

Bayes, Phil. Trans. Roy. Soc., **53**, 370 (1763)

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model comparison

$$\frac{p(H_x|\mathbf{D},\mathbf{I})}{p(H_y|\mathbf{D},\mathbf{I})} = \frac{p(\mathbf{D}|H_x,\mathbf{I})}{p(\mathbf{D}|H_y,\mathbf{I})} \times \frac{p(H_x|\mathbf{I})}{p(H_y|\mathbf{I})}$$

Pullen and Morris, PLOS ONE **9**, e88419 (2014)

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parity between the hypotheses

$$\frac{p(H_x|\mathbf{D},\mathbf{I})}{p(H_y|\mathbf{D},\mathbf{I})} = \frac{p(\mathbf{D}|H_x,\mathbf{I})}{p(\mathbf{D}|H_y,\mathbf{I})}$$

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possibly multi-dimensional

$$p(\mathbf{D}|H,\mathbf{I}) = \iint_{\mathbf{R}} \mathcal{L}(\mathbf{X})p(\mathbf{X}|H,\mathbf{I})\mathrm{d}^M\mathbf{X}$$

🖥💵🖥💵🖥💵🖥💵🖥💵🖥

Sivia and Skilling, *Data Analysis: A Bayesian Tutorial*, Oxford University Press, Oxford (2006)

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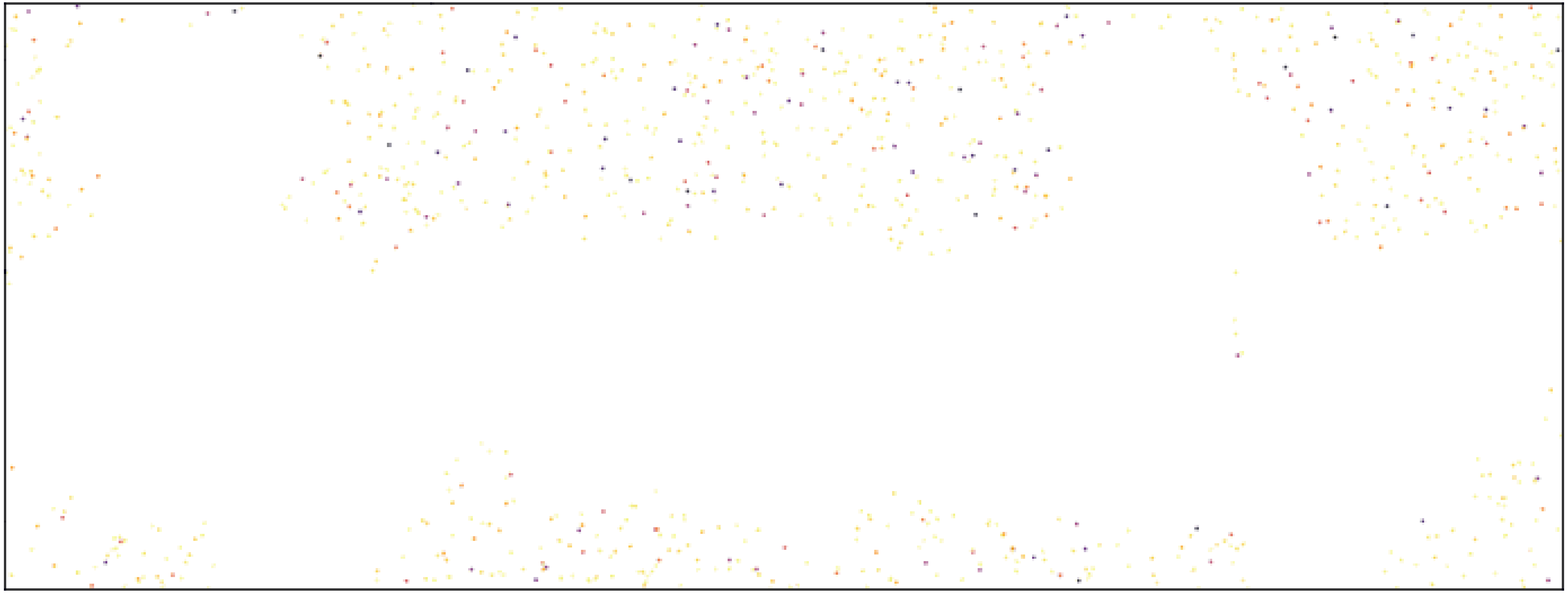

Dynamic nested sampling using dynesty

Higson *et al.*, arXiv: 1704.03459; Speagle, arXiv: 1904.02180

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nested sampling

- Draw K "live points" from the prior
- Get natural logarithm of likelihood at each live point
- Kill the live point with the lowest likelihood
- Draw new live point from prior subject to the constraint
$$\mathcal{L}_{i+1} \geq \mathcal{L}_i$$
- Continue until stopping criteria is reached

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evidence evaluation

$$p(\mathbf{D}|H, \mathbf{I}) \approx \sum_{i=1}^{N} \frac{\mathcal{L}_{i-1} + \mathcal{L}_i}{2} (X_{i-1} - X_i)$$

$$\ln X_i \approx -\frac{i \pm \sqrt{i}}{K}$$

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dynamic nested sampling

- The number of live points is dynamically allocated
- This allows the algorithm to adapt to the shape of the evidence distribution in real time
- Leading to improved accuracy and efficiency

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Bayes' factor

$$\ln(B_{x,y}) = \ln\left[\frac{p(\mathbf{D}|H_x,\mathbf{I})}{p(\mathbf{D}|H_y,\mathbf{I})}\right] = \begin{cases} > 0, \text{ prefer model } x \\ \approx 0, \text{ no preference} \\ < 0, \text{ prefer model } y \end{cases}$$

Sivia *et al.*, Physica D, **66**, 234 (1993)

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Bayesian model comparison

- This offers a clear, robust, and scalable method for comparison of reflectometry models
  - The evidence is *data-dependent*, so can compare different data quantities
  - Not the first to apply Bayesian model comparison to neutron reflectometry

Sivia *et al.*, Physica B, **173**, 121 (1991); Geoghagen *et al.*, Thin Solid Films, **53**, 825 (1996); Sivia and Webster, Physica B, **248**, 327 (1998)

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what did we do?

- Apply this Bayesian method to the three Campbell models
- Investigate the effect of different isotopic contrasts
- Show (mathematically) what everyone already knew

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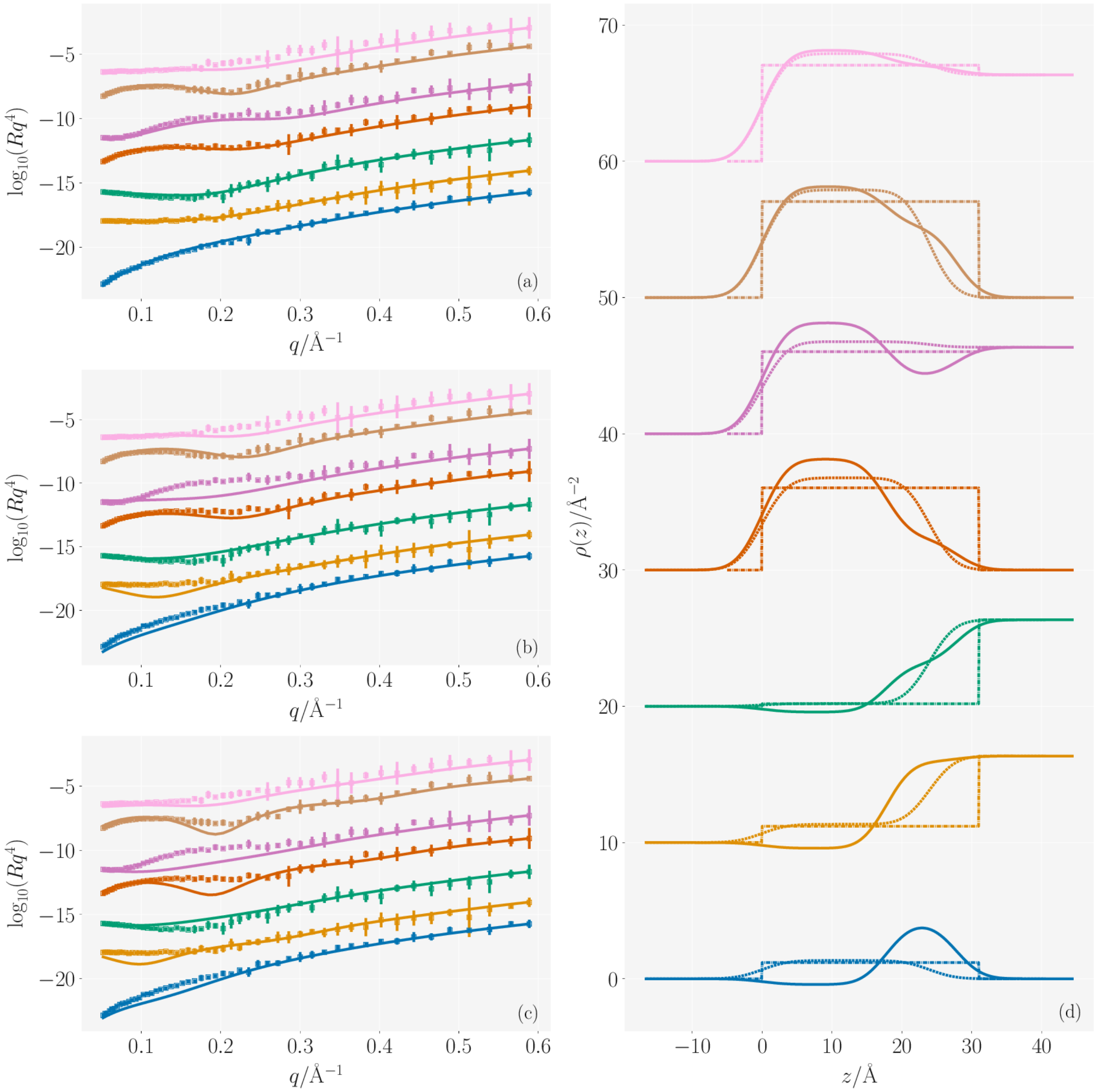

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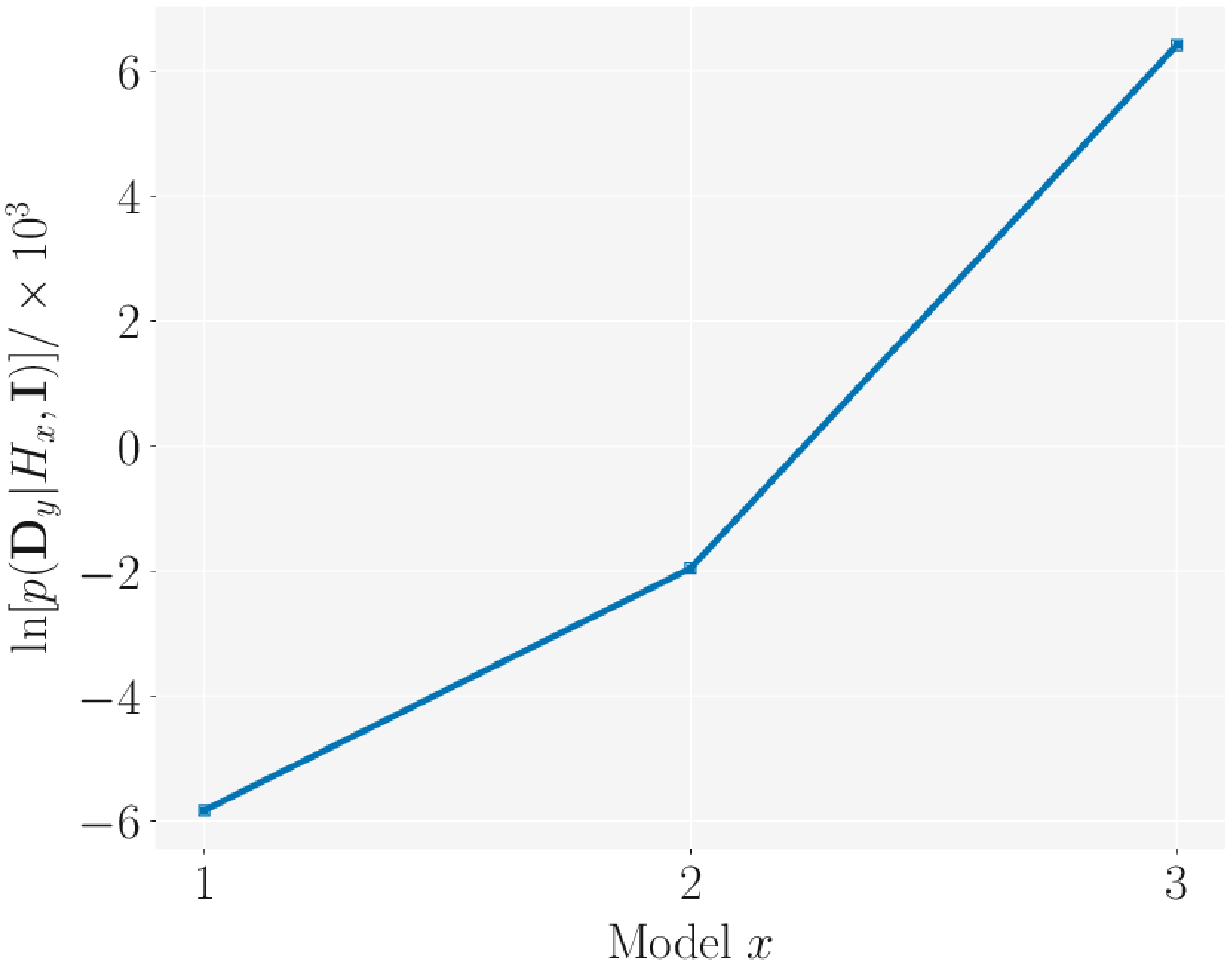

Agrees well with the Campbell *et al.* result

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but getting seven contrasts of neutron data isn't easy

- usually expensive and synthetically difficult
- most of the time only a few contrasts are used

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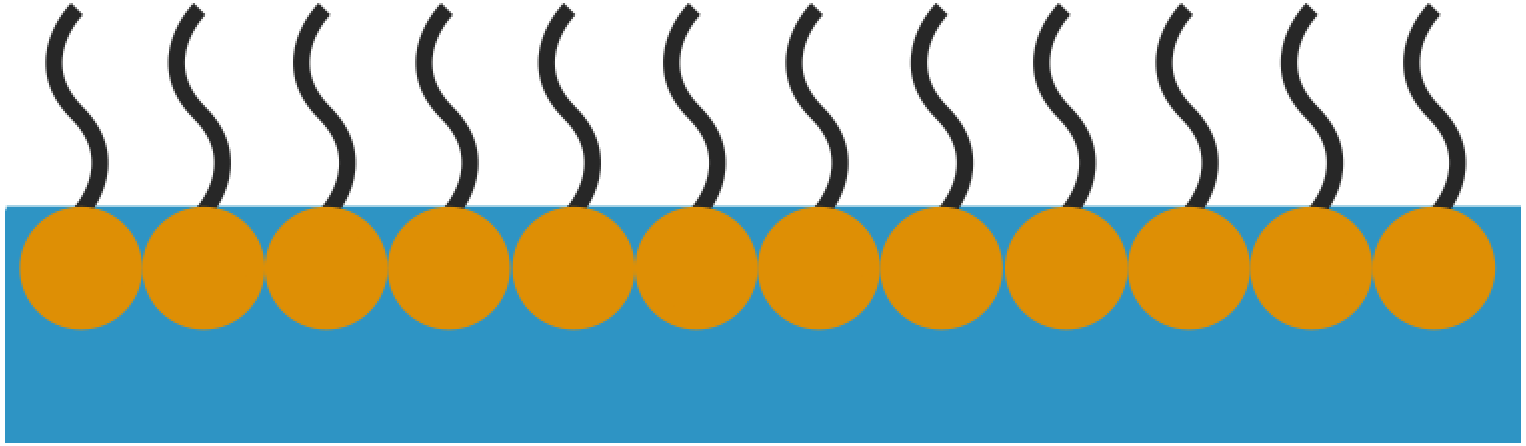
the most common contrasts



- tail-deuterated with D_2O and ACMW and fully-hydrogenated with D_2O

Pabois *et al.*, J. Colloid Interface Sci., **556**, 266 (2019); Ortiz-Collazos *et al.*, BBA - Biomembranes, **1861**, 182994 (2019)

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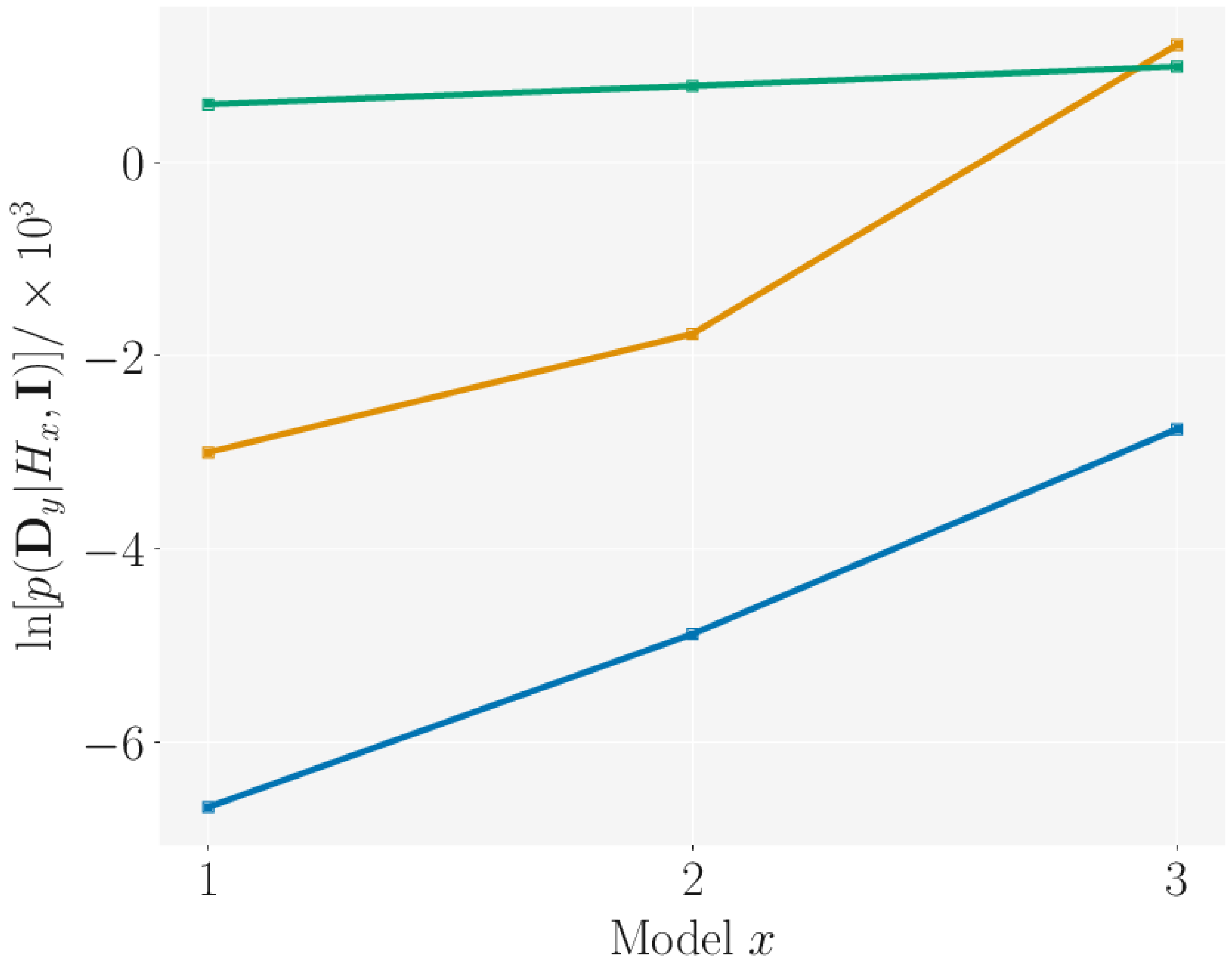

Fewer contrasts, less relative evidence

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is there a combination where model 3 doesn't have the most evidence?

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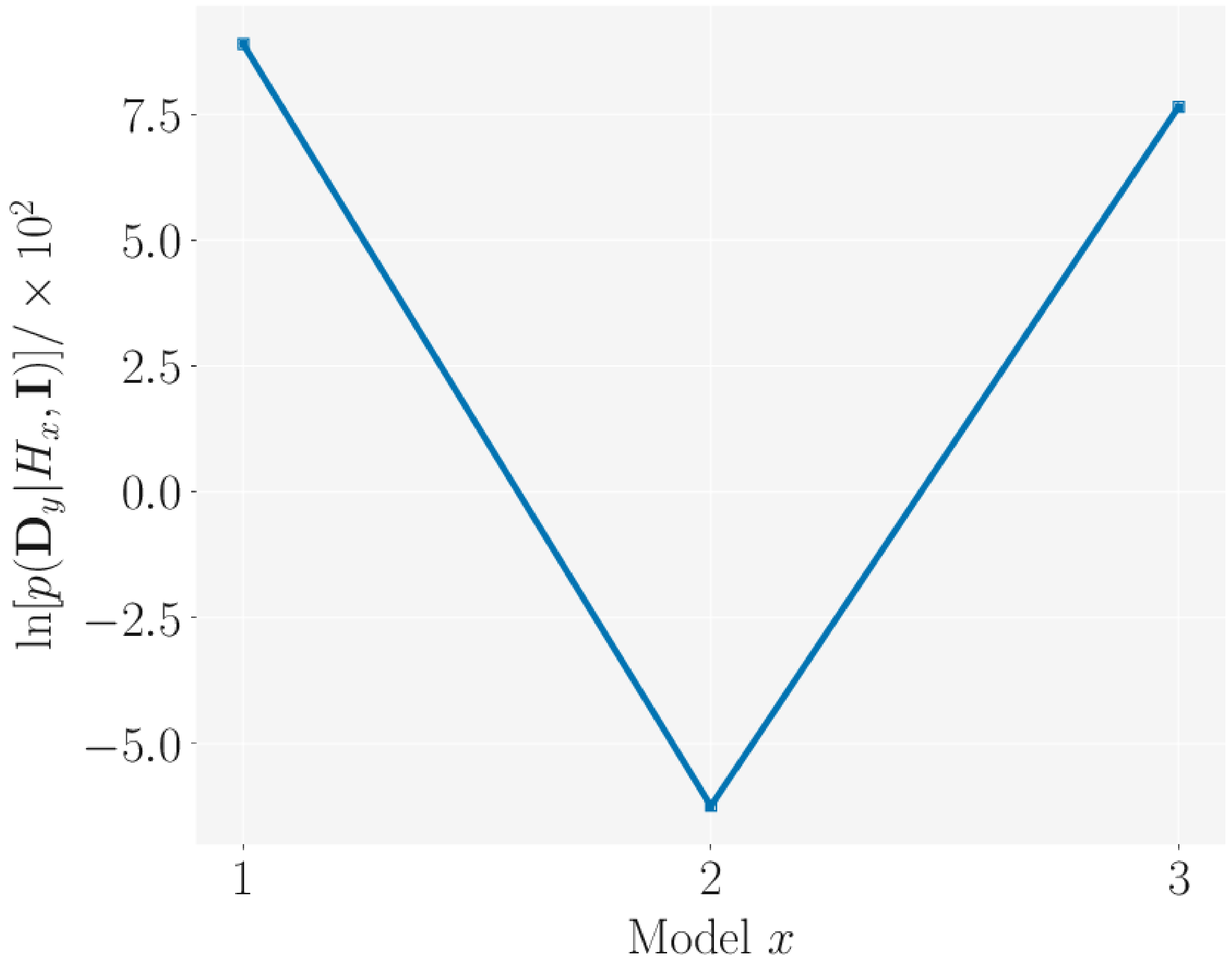

Fully-deuterated molecules on D_2O and ACMW and fully hydrogenated molecules on D_2O

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consider the evidence for different models

- If an experimentalist wants to use a model of lesser evidence, important to accept that this is based on a qualitative understanding of the system

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conclusions

- Statistically rigorous method for the comparison of relative model evidence
  - *Known knowledge* is usually true
  - Fewer contrasts, less confidence
- Lots of nice work on this possible in the future

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thanks for listening

Penny and Sadie, good dogs