Simulation-Informed Bayesian Inference Improves Understanding of Molecular Motion From Neutron Scattering

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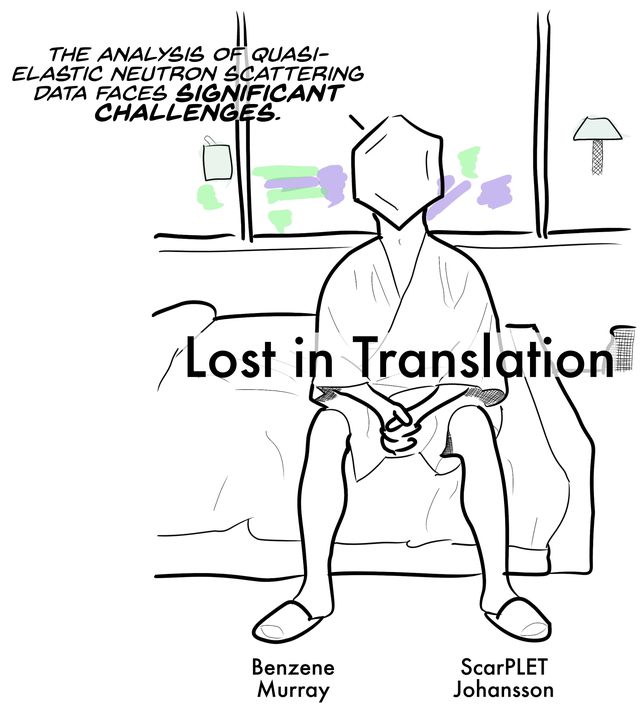
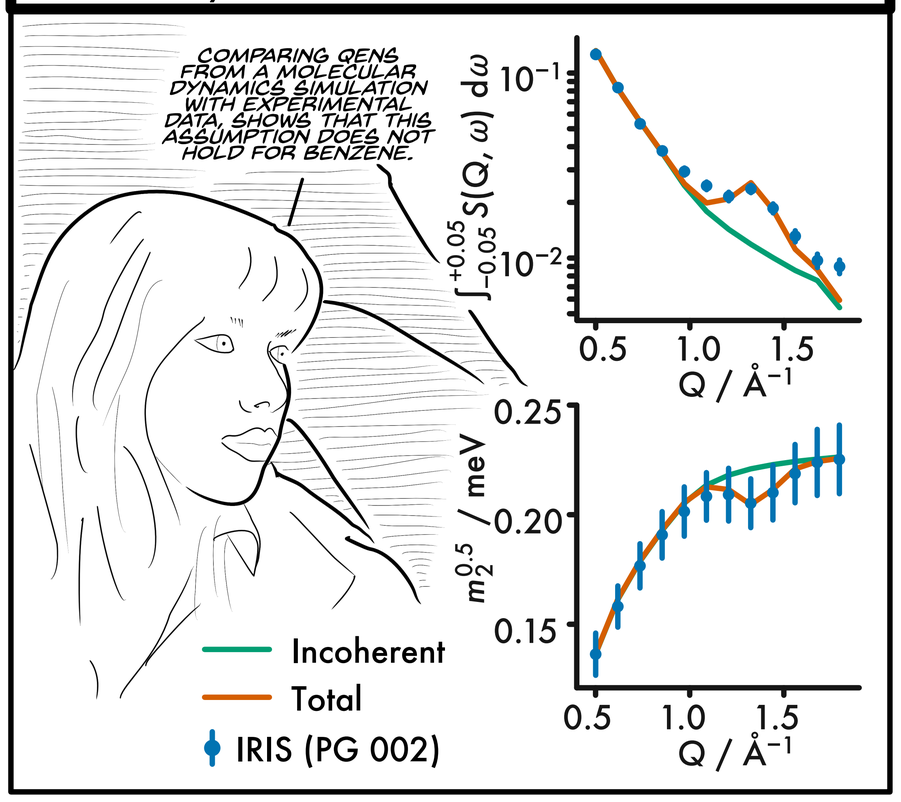
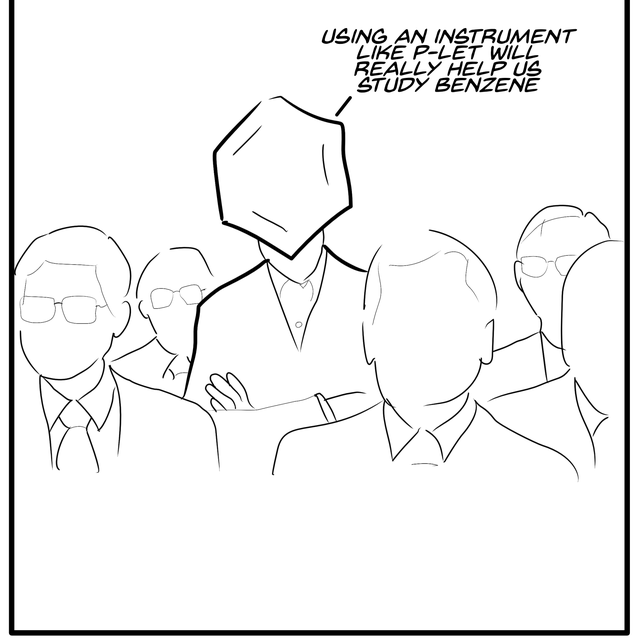


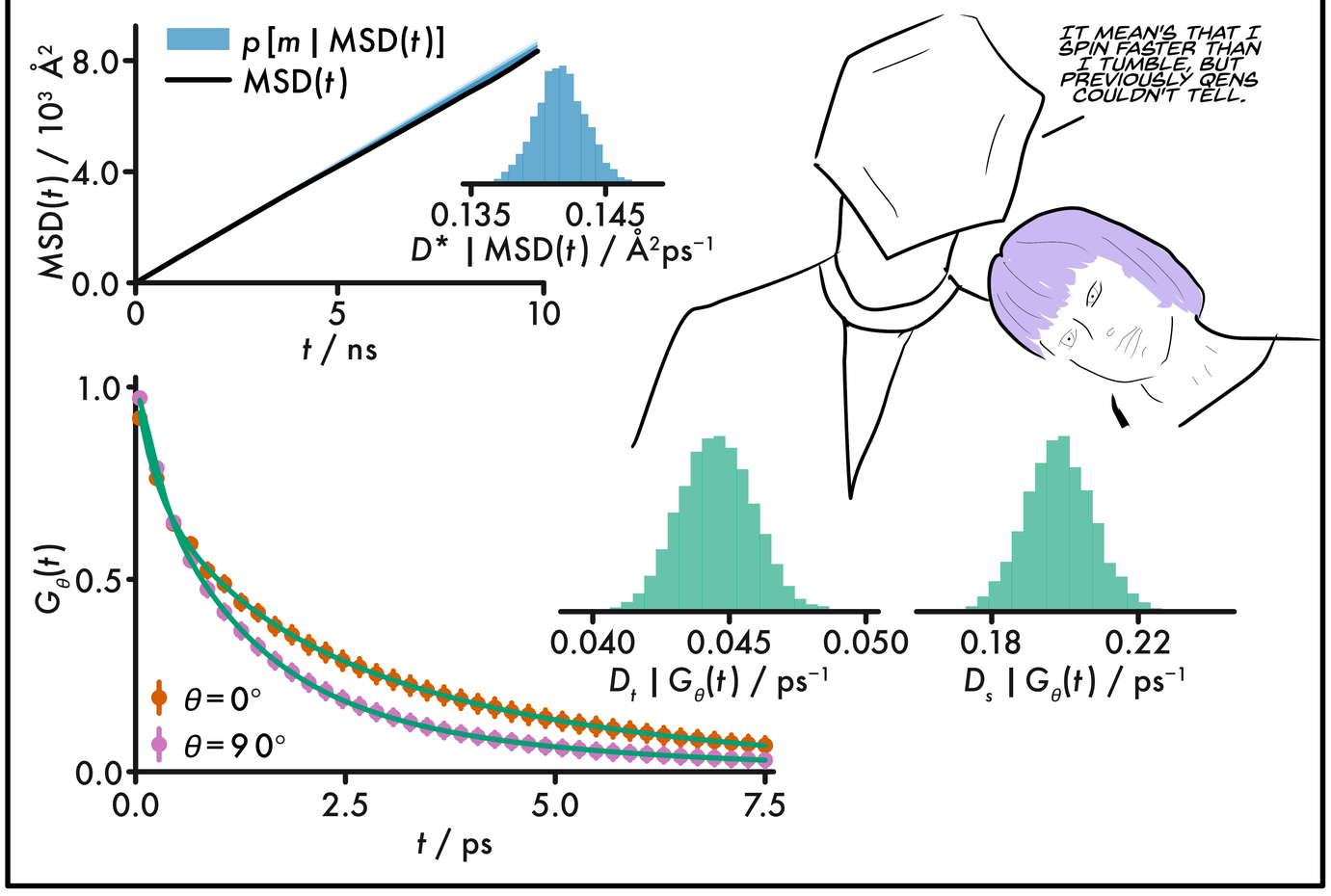
The analysis of standard QENS makes **a significant assumption** about the observed data; when the system is hydrogenous, the observed signal is only the incoherent dynamic structure factor.



Therefore, for many experimental systems, e.g., liquid benzene, it is necessary to either model the coherent signal or **measure QENS with polarisation analysis**.



Often QENS models represent oversimplified descriptions of molecular dynamics. Classical simulations of benzene showed a single Fickian regime in translation, but **complex anisotropic rotation**.



It was necessary to construct a new QENS model, that describes Fickian translation and anisotropic rotation and is **constrained over Q**.

Translation

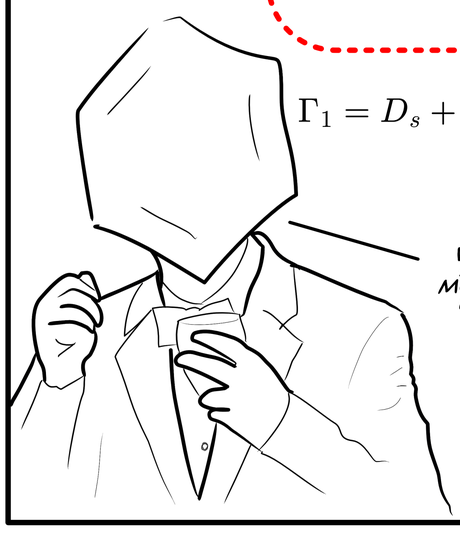
$$S_{inc}(Q,\omega) = e^{-Q^2\langle u^2\rangle/3}\left[\frac{1}{\pi}\frac{D^*Q^2}{\omega^2+(D^*Q^2)^2}\right] \otimes j_0^2(QR)\,\delta(\omega)$$

$$+\,3\,j_1^2(QR)\left[\frac{1}{\pi}\frac{\Gamma_1}{\omega^2+\Gamma_1^2}\right]$$

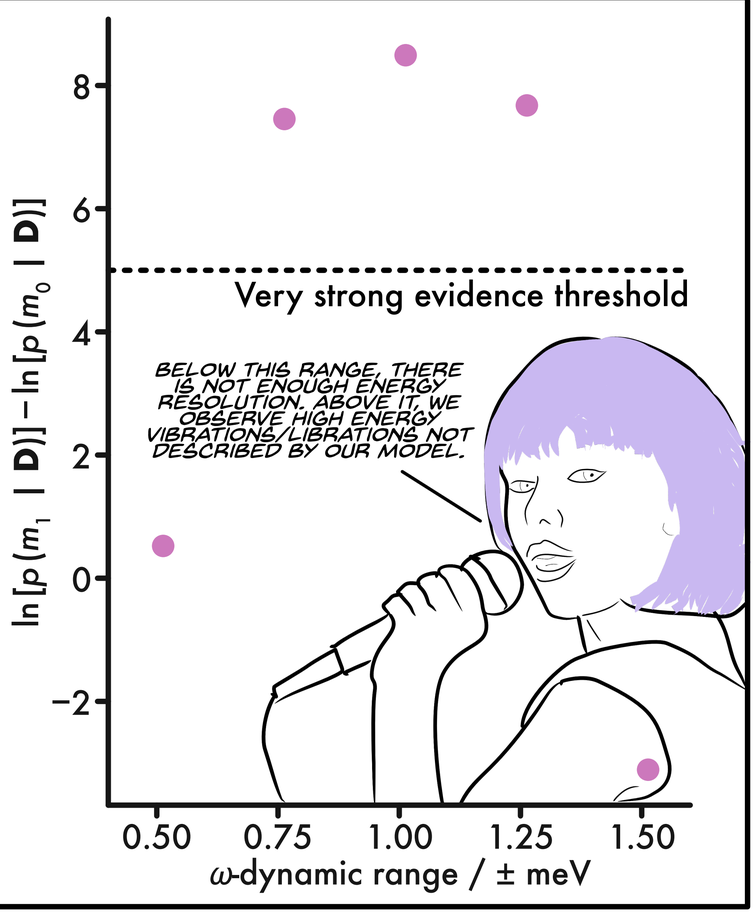
$$+\,5\,j_2^2(QR)\left[\frac{1}{4}\frac{1}{\pi}\frac{\Gamma_2}{\omega^2+\Gamma_2^2}+\frac{3}{4}\frac{1}{\pi}\frac{\Gamma_3}{\omega^2+\Gamma_3^2}\right]$$

Rotation

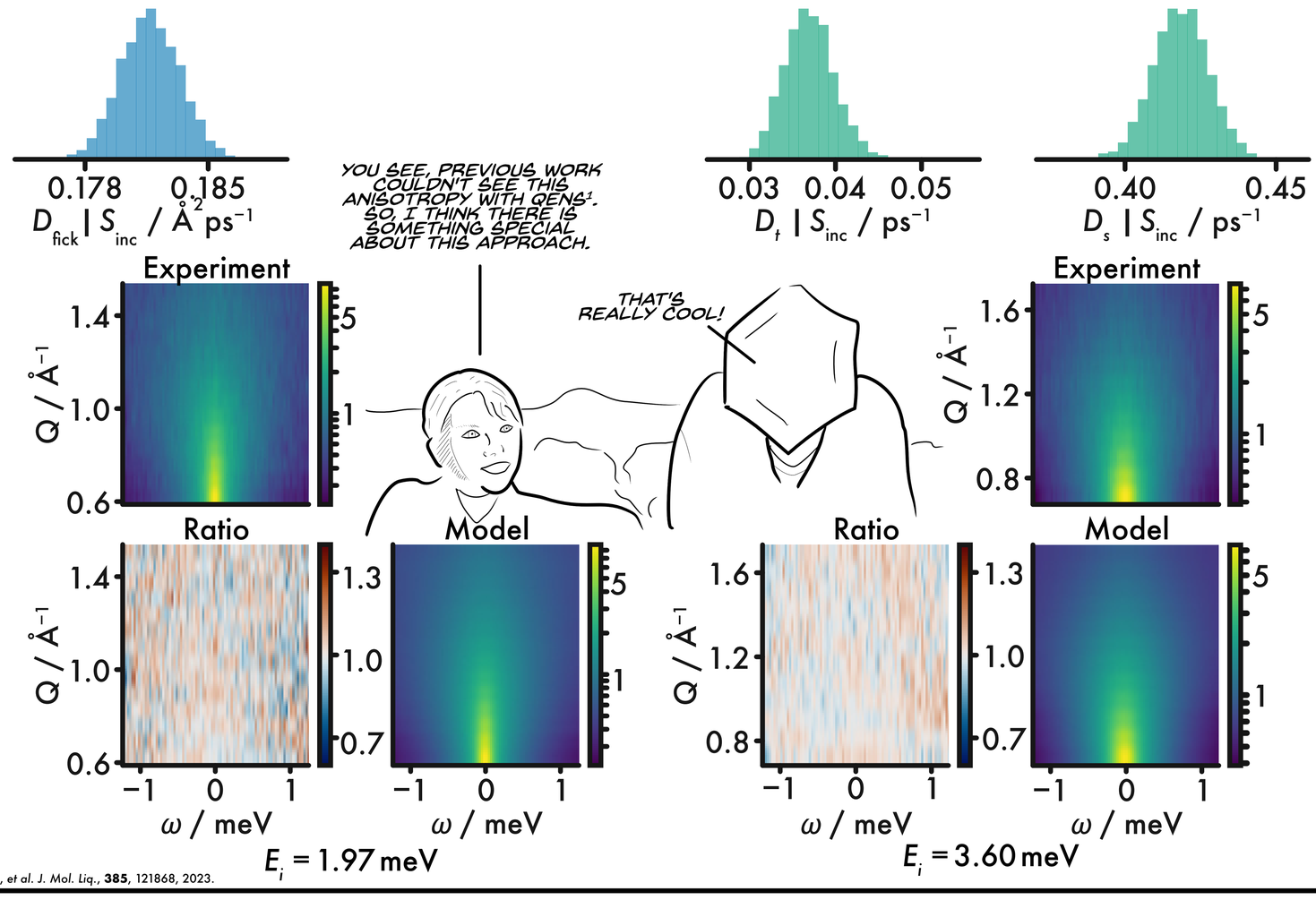
$$\Gamma_1 = D_s + D_t \quad \Gamma_2 = 6D_t \quad \Gamma_3 = 4D_s + 2D_t$$



Bayesian model selection allows us to **compare different models**. Applying this to our QENS from simulation shows strong evidence for the anisotropic model, if the ω-dynamic range is between ±0.75 and ±1.25 meV.



The anisotropic model, given p-QENS data from LET, also has greater evidence for the *more complex*, anisotropic model. The ratio of benzene spinning to tumbling was found to be around 11.



1. Kikuchi, *et al. J. Mol. Liq.*, **385**, 121868, 2023.

Read the paper on *arXiv*:2603.06080.

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