Accurate Estimation of Self- and Collective-Motion from Molecular Dynamics Simulation

PAQMAN — 2025-07-03

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scams-research.github.io
(he/him)

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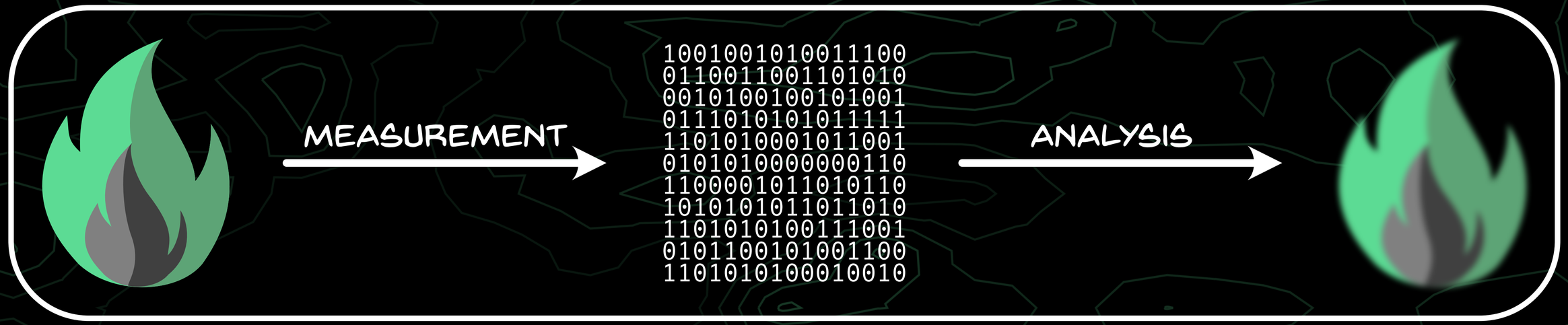
THE MEASUREMENT PROCESS IS A **NOISY CHANNEL**; INFORMATION IS LOST. THE ROLE OF ANALYSIS IS TO TRY AND RECONSTRUCT THE PHYSICAL OBSERVABLE.



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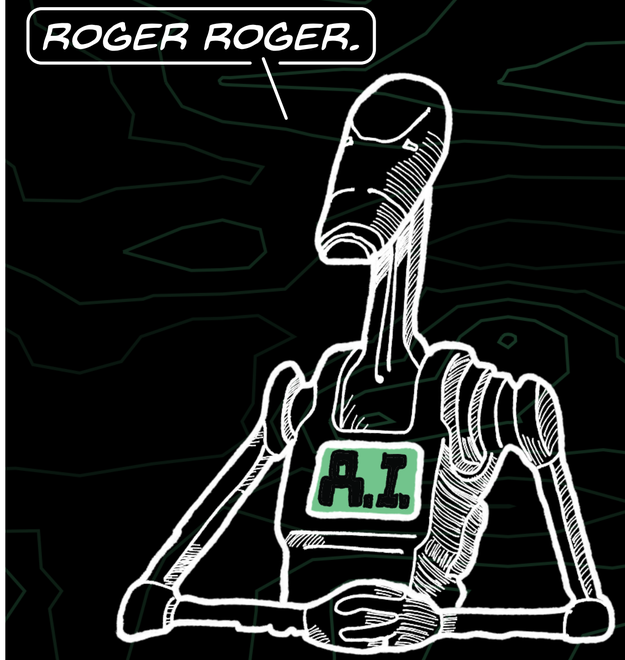
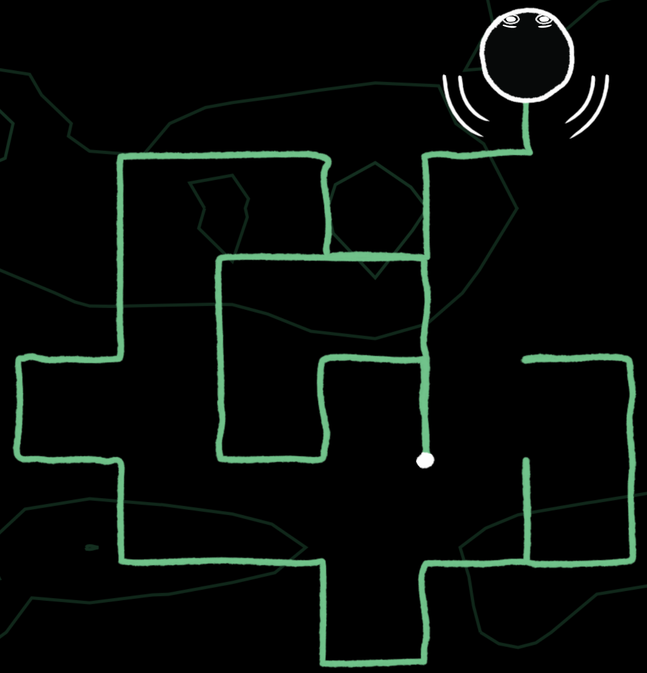
DIFFUSION PLAYS AN IMPORTANT ROLE IN ENERGY MATERIALS AND INDUSTRIAL PROCESSES.

AN UNDERSTANDING OF DIFFUSION CAN FACILITATE RATIONAL DESIGN, LEVERAGING ARTIFICIAL INTELLIGENCE.



PLUS, IT CAN BE FUN TO WORRY ABOUT RANDOM WALKS.



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ARRHENIUS MODELLING CAN BE USED FOR DIFFUSION...

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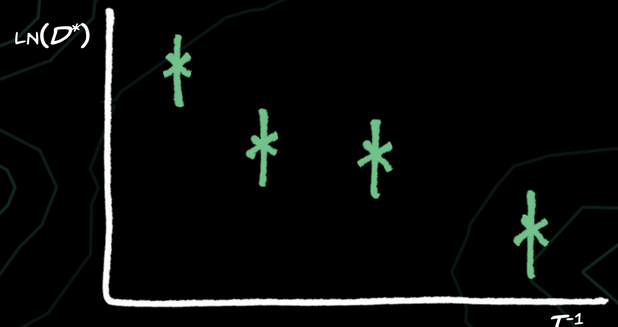
ARRHENIUS MODELLING CAN BE USED FOR DIFFUSION...

...AND WITHOUT ERROR BARS RESULTS ARE MEANINGLESS.



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ARRHENIUS MODELLING CAN BE USED FOR DIFFUSION...

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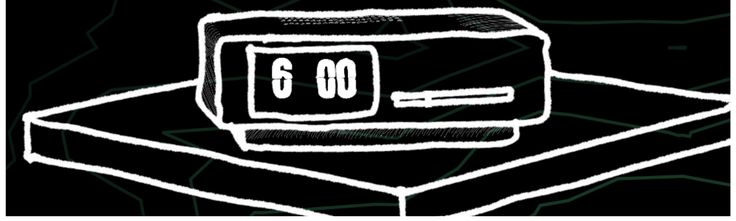
REPEATING SIMULATIONS CAN BECOME TEDIOUS...

THEN PUT YOUR LITTLE HAND IN MINE...



THEN PUT YOUR LITTLE HAND IN MINE...

THEN PUT YOUR LITTLE HAND IN MINE... xN

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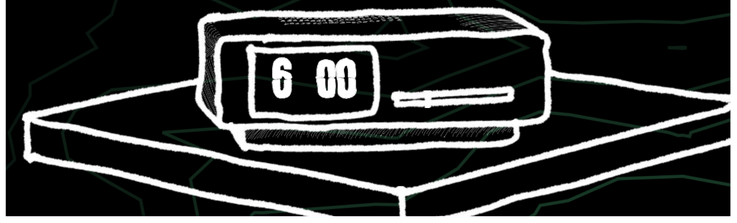

THEN PUT YOUR LITTLE HAND IN MINE...

THEN PUT YOUR LITTLE HAND IN MINE... xN

...SO WE WANT THE BEST WAY TO MAXIMISE THE ***PRECISION*** OF OUR ESTIMATE AND THE ***ACCURACY*** OF THE VARIANCE ESTIMATE.

IDEALLY, FROM A SINGLE MEASUREMENT.

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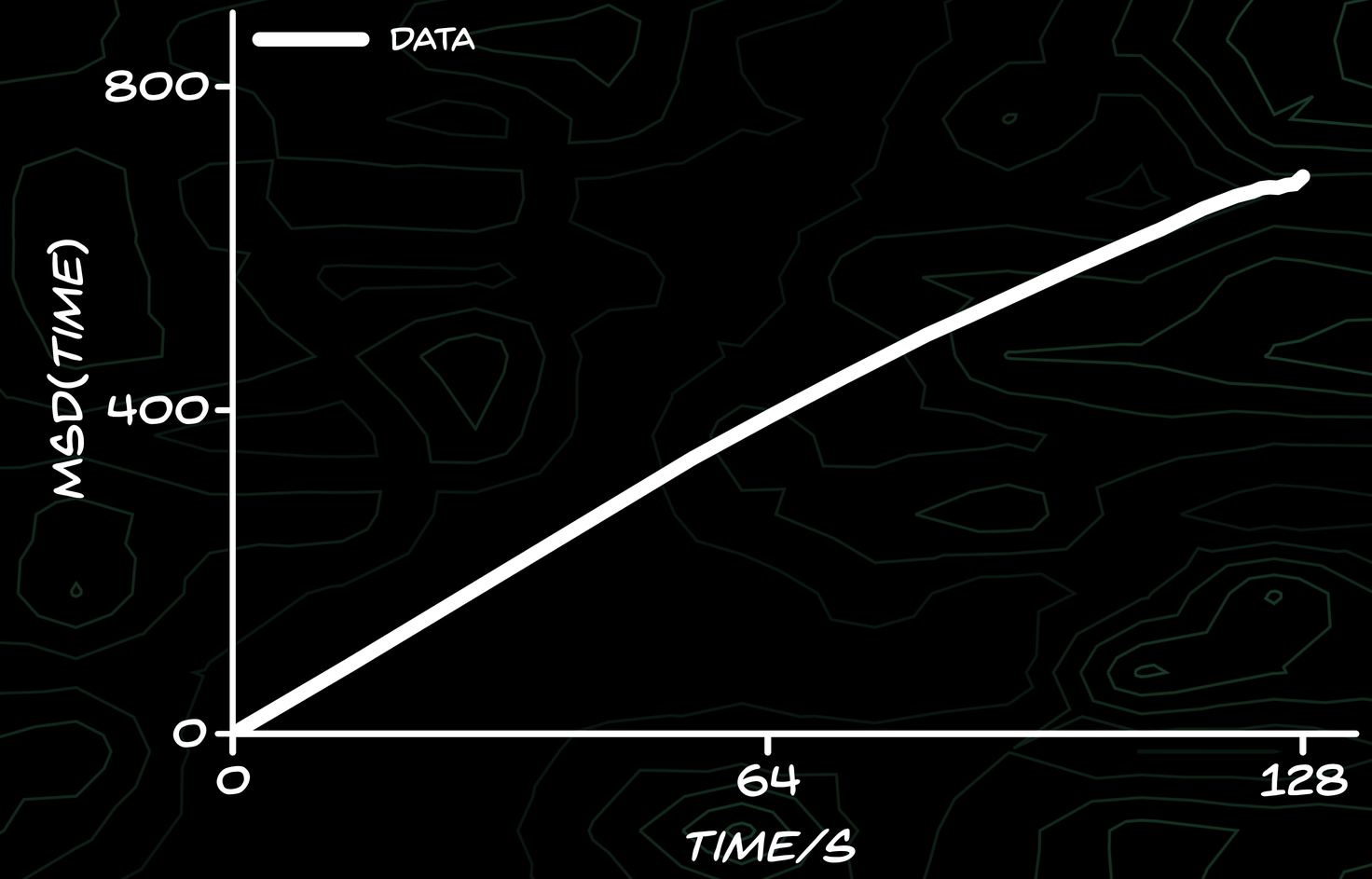

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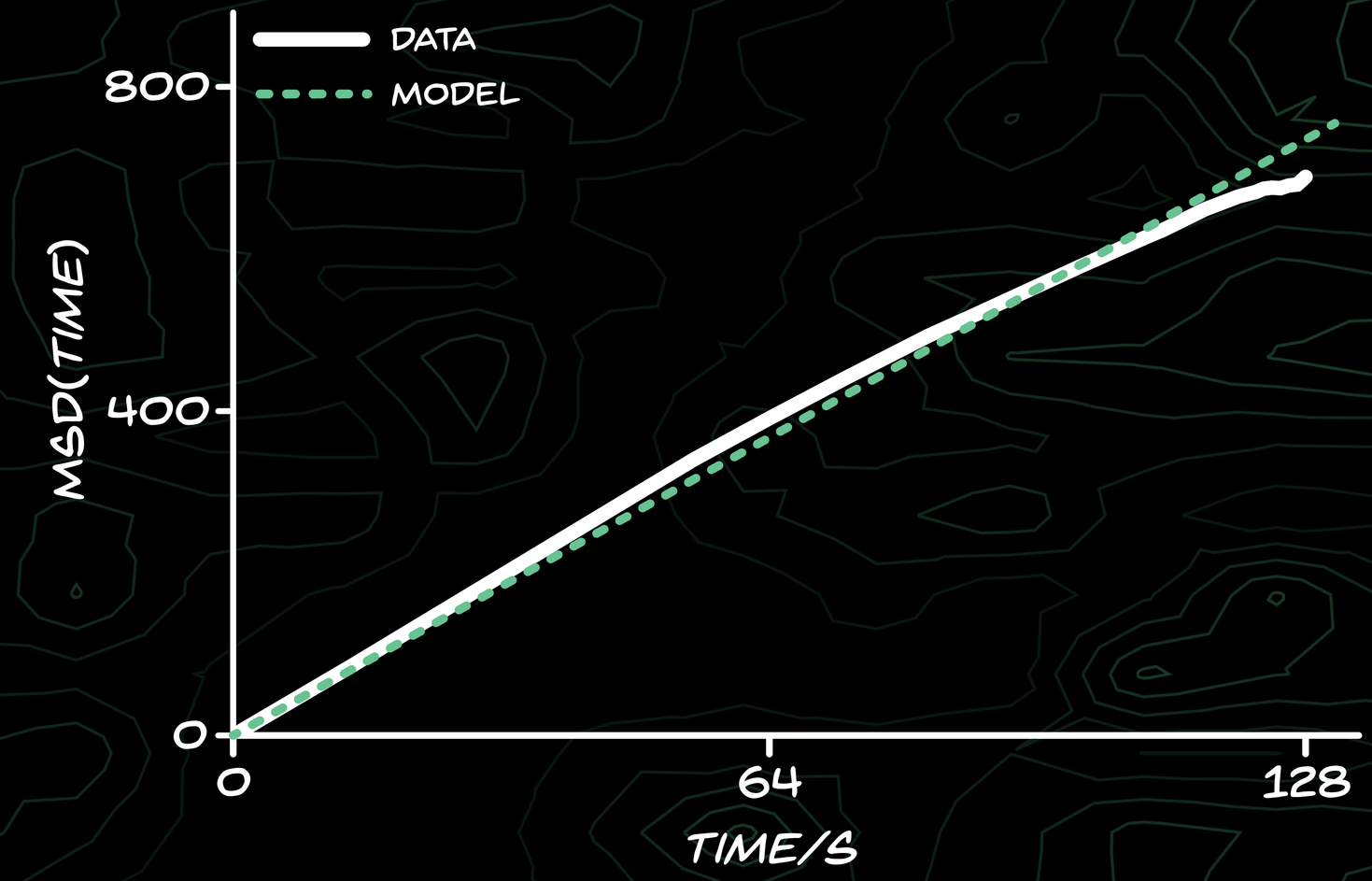

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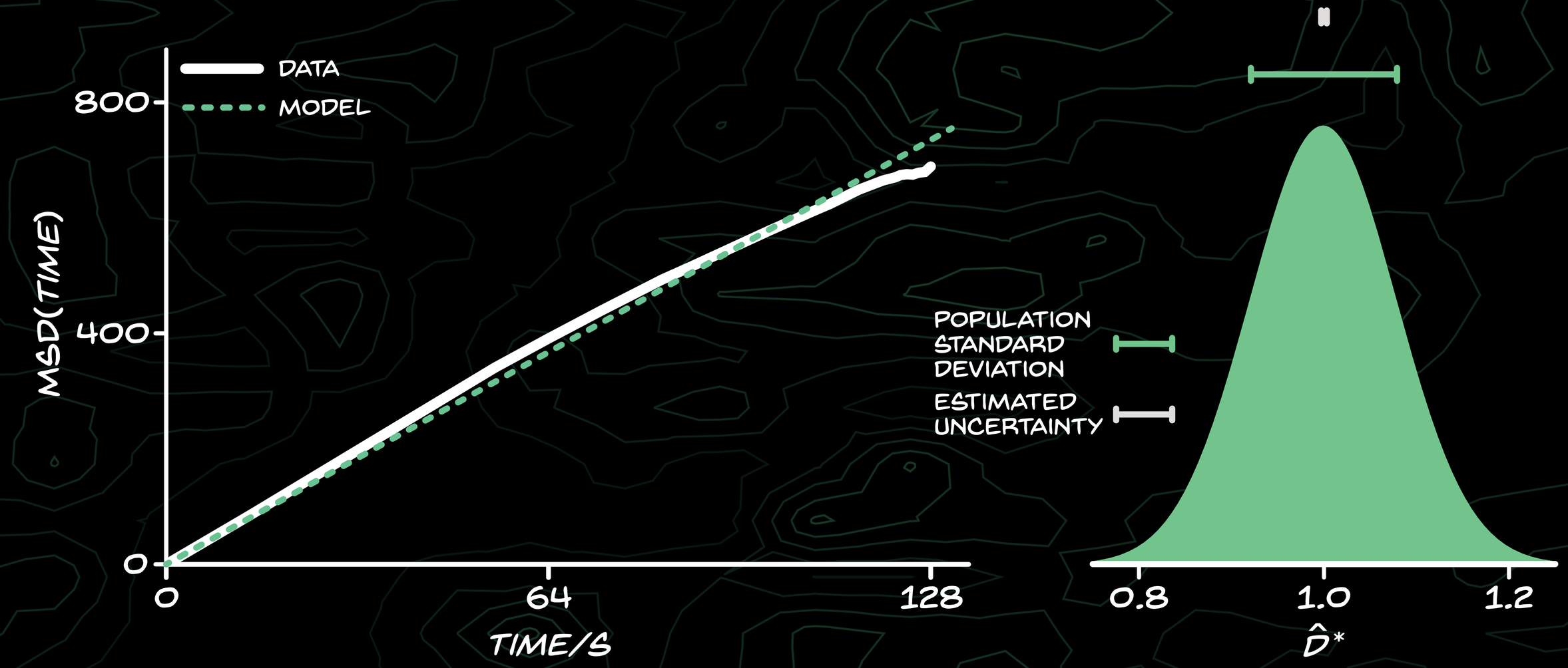

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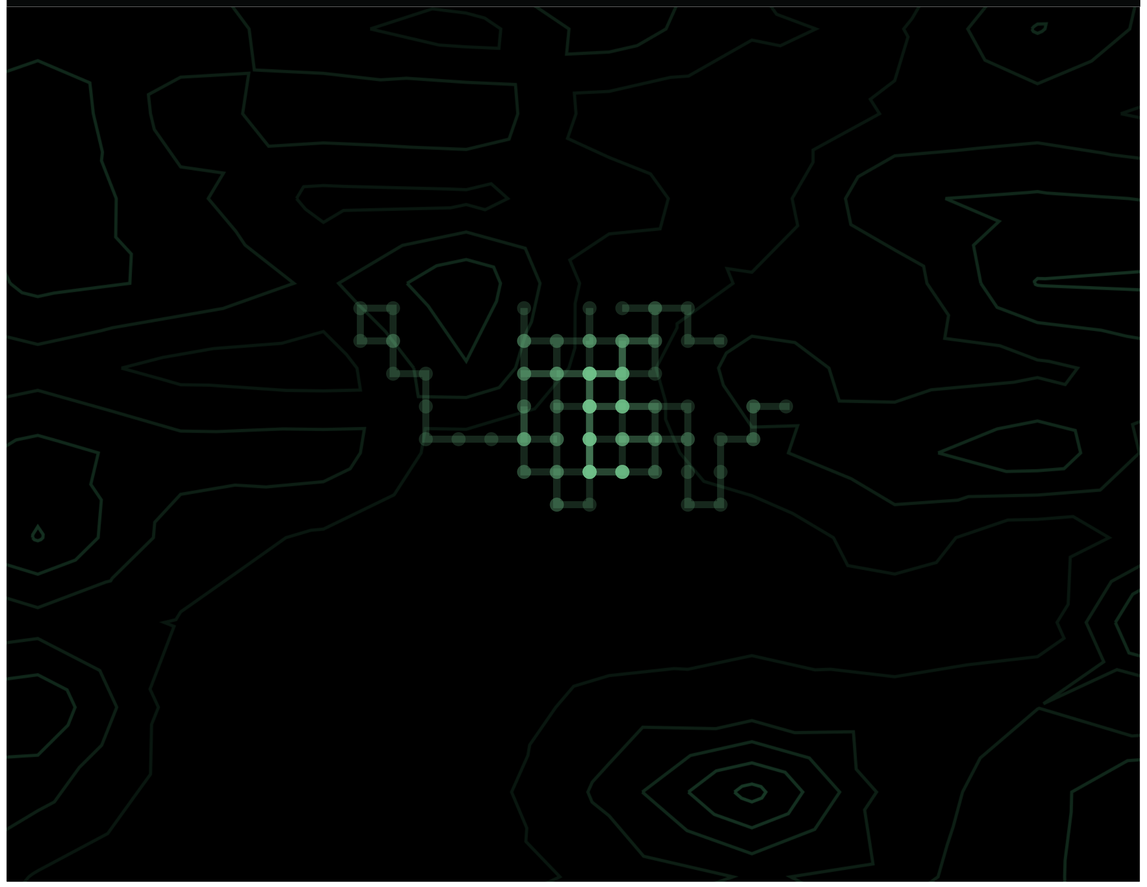
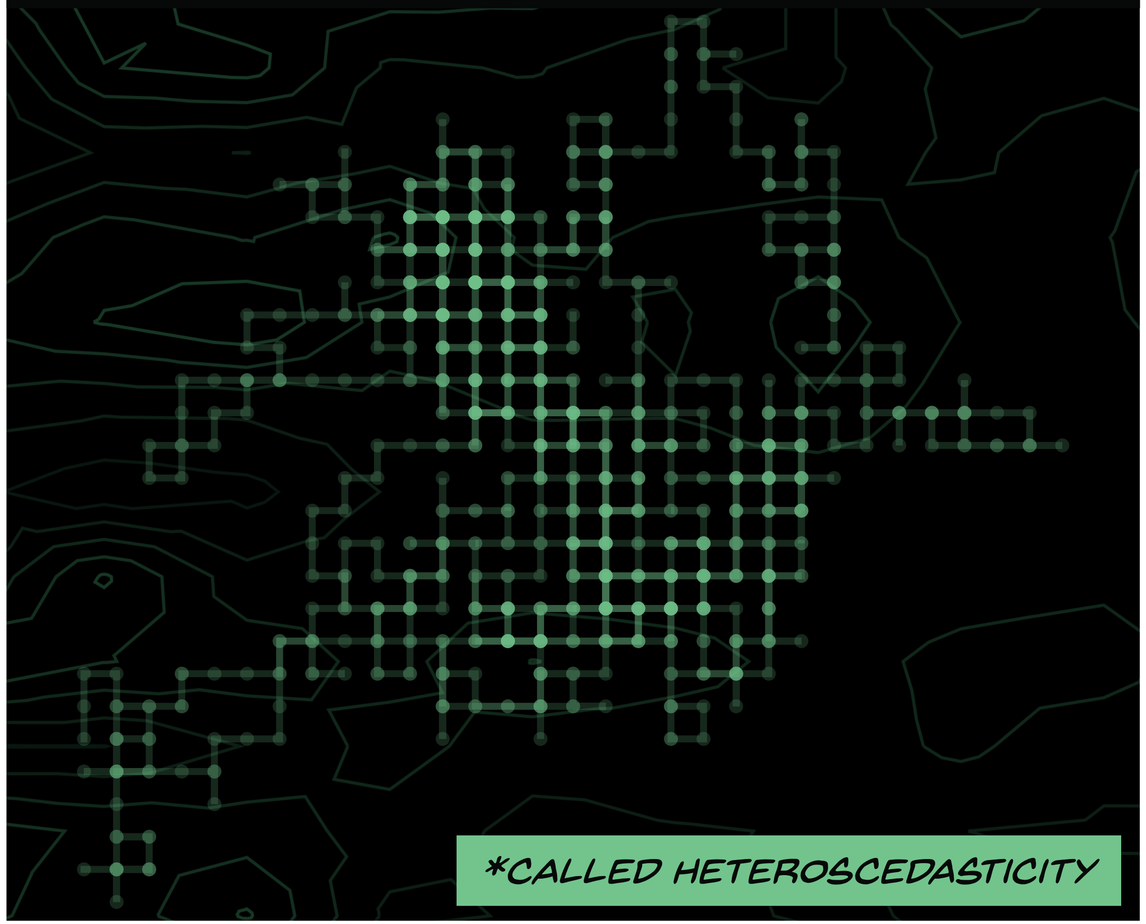
AFTER 16 2D RANDOM WALK STEPS, THERE ARE A RANGE OF VALUES OF DISPLACEMENT THAT CAN BE REACHED....



...AFTER LONGER PERIODS OF TIME, THAT RANGE (THE VARIANCE) INCREASES*.



*CALLED HETEROSCEDASTICITY

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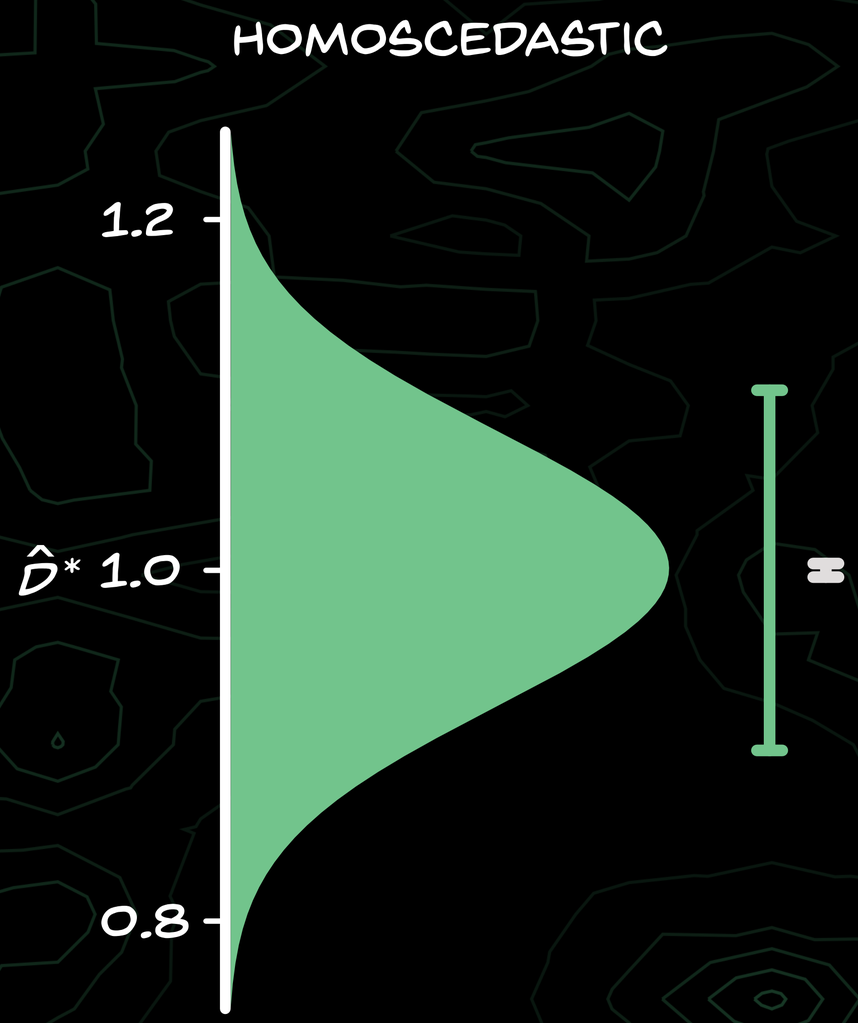

POPULATION STANDARD DEVIATION

ESTIMATED UNCERTAINTY

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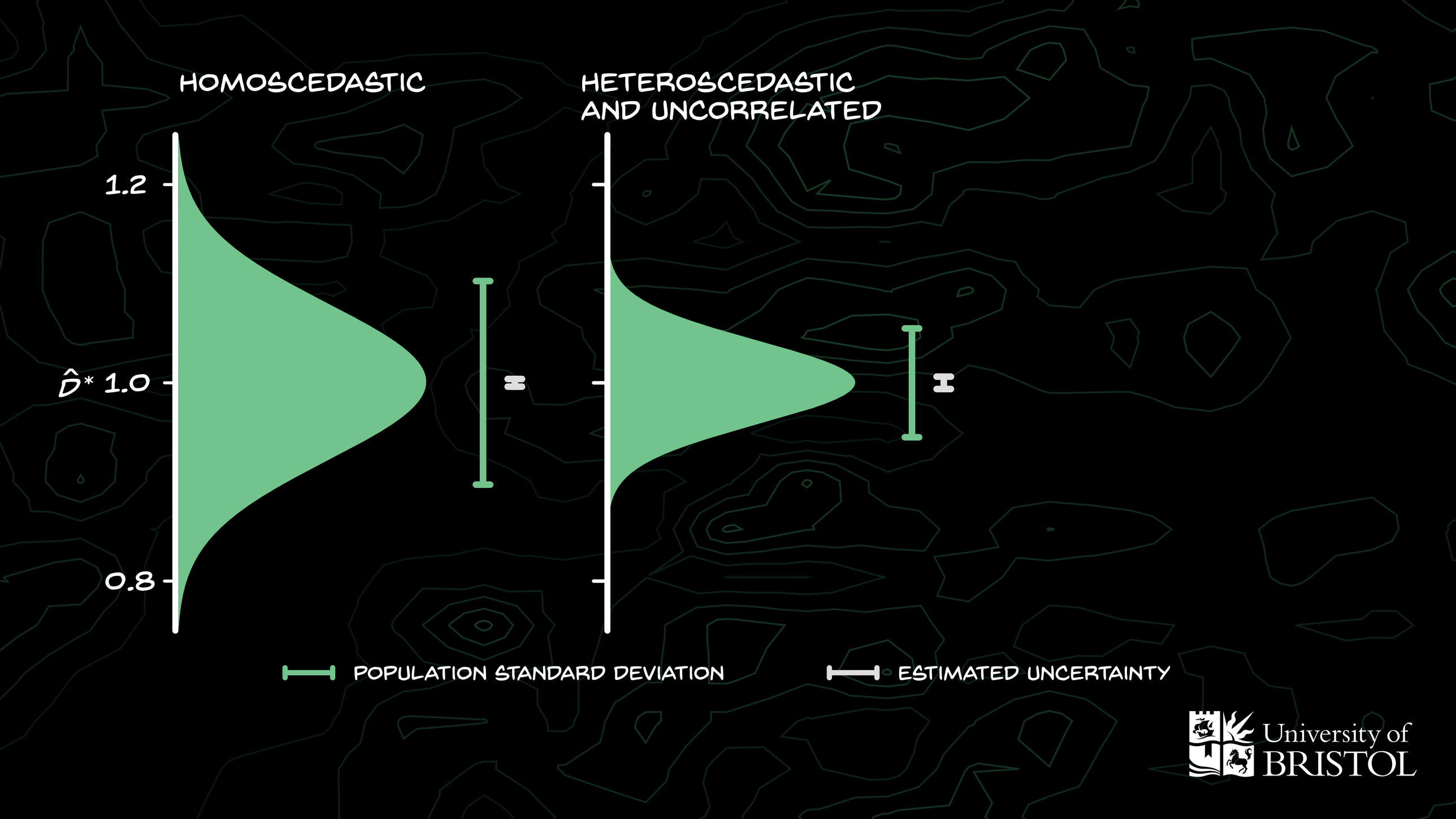

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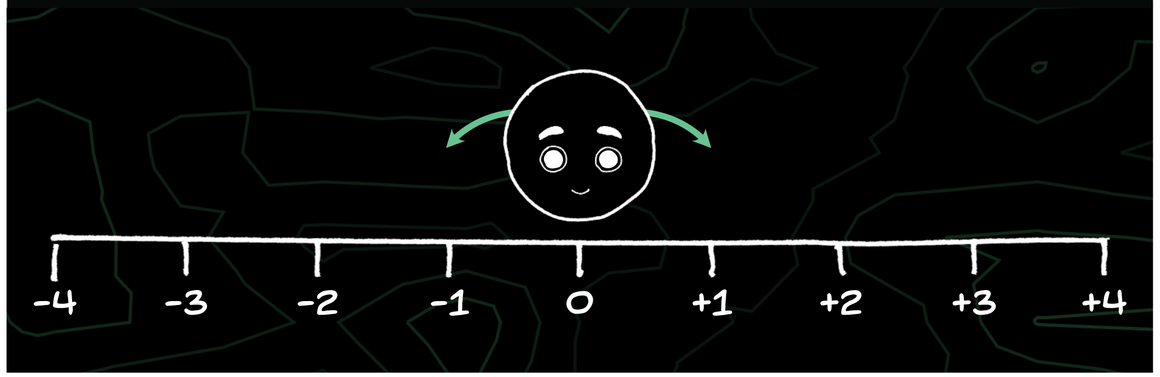
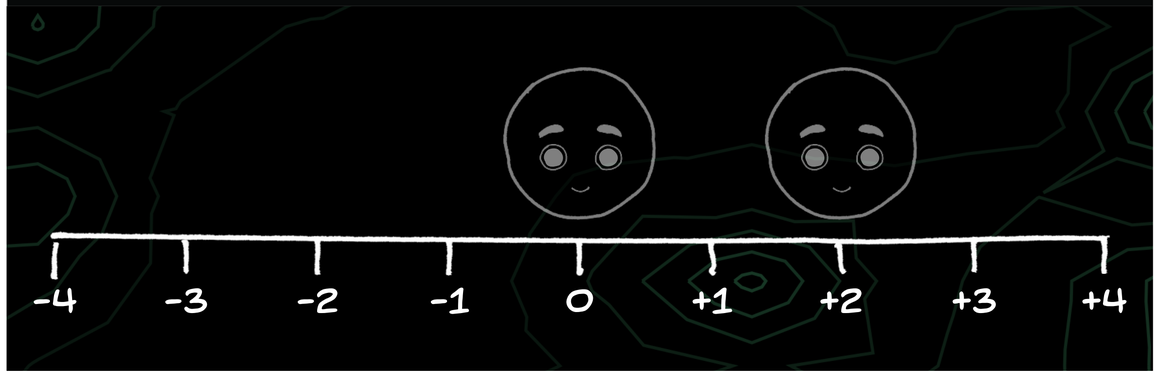
IF A PARTICLE ON A 1D RANDOM WALK WITH A STEP SIZE OF 1...

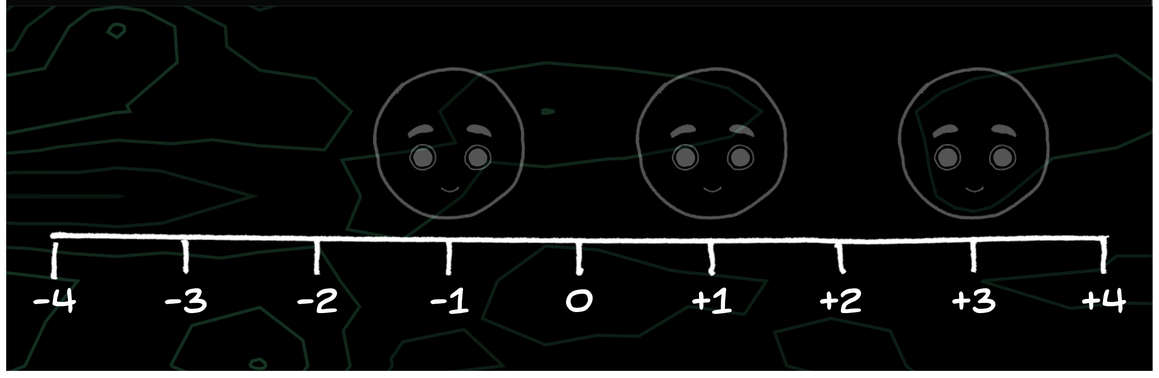


...AND AFTER 5 STEPS, IT GETS TO THE POSITION +1.

THEN AFTER 6 STEPS, IT COULD ONLY BE AT POSITIONS OF 0 OR +2...



...AND AFTER 7 STEPS, IT CAN ONLY BE AT POSITIONS -1, +1, OR +3.



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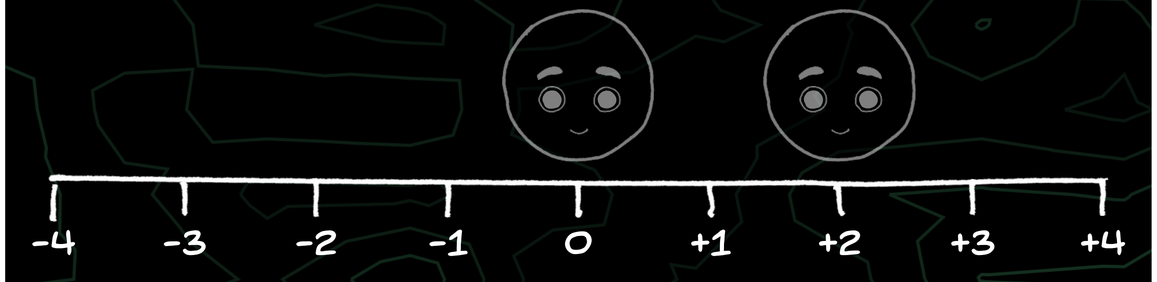

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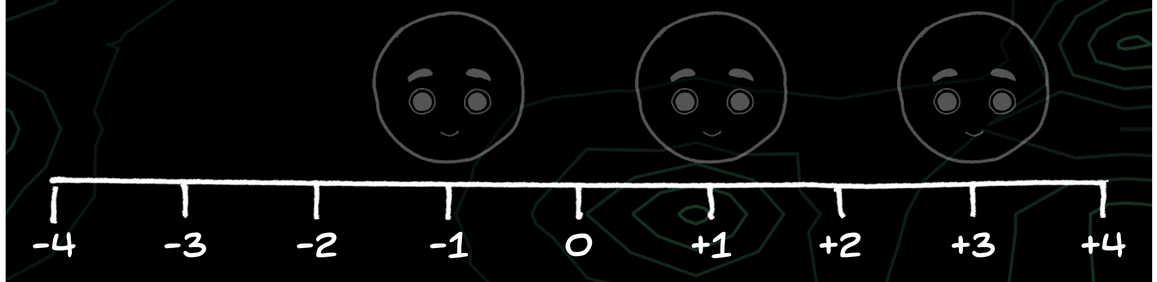
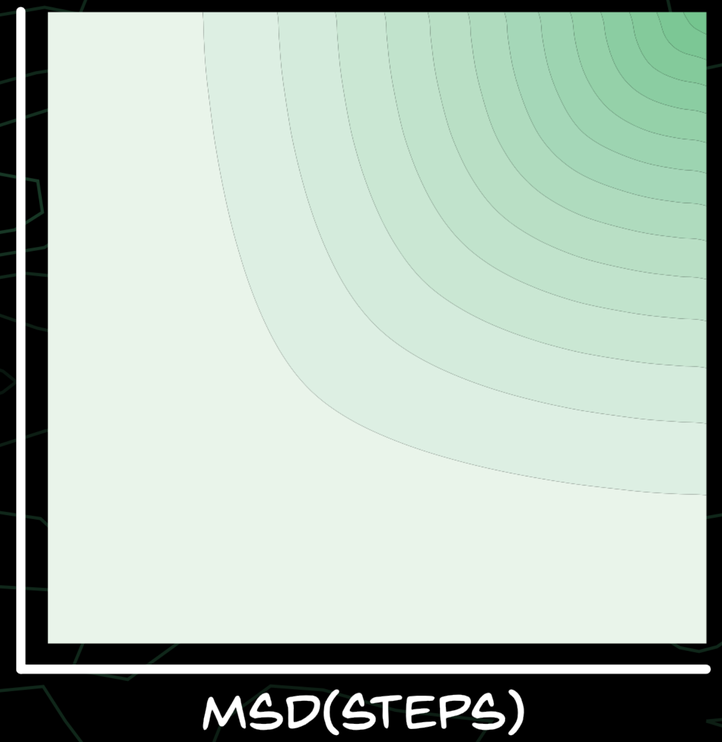


THE AMOUNT THAT THE DIFFERENT TIME POINTS INFLUENCE EACH OTHER IS DEFINED IN THE COVARIANCE MATRIX.



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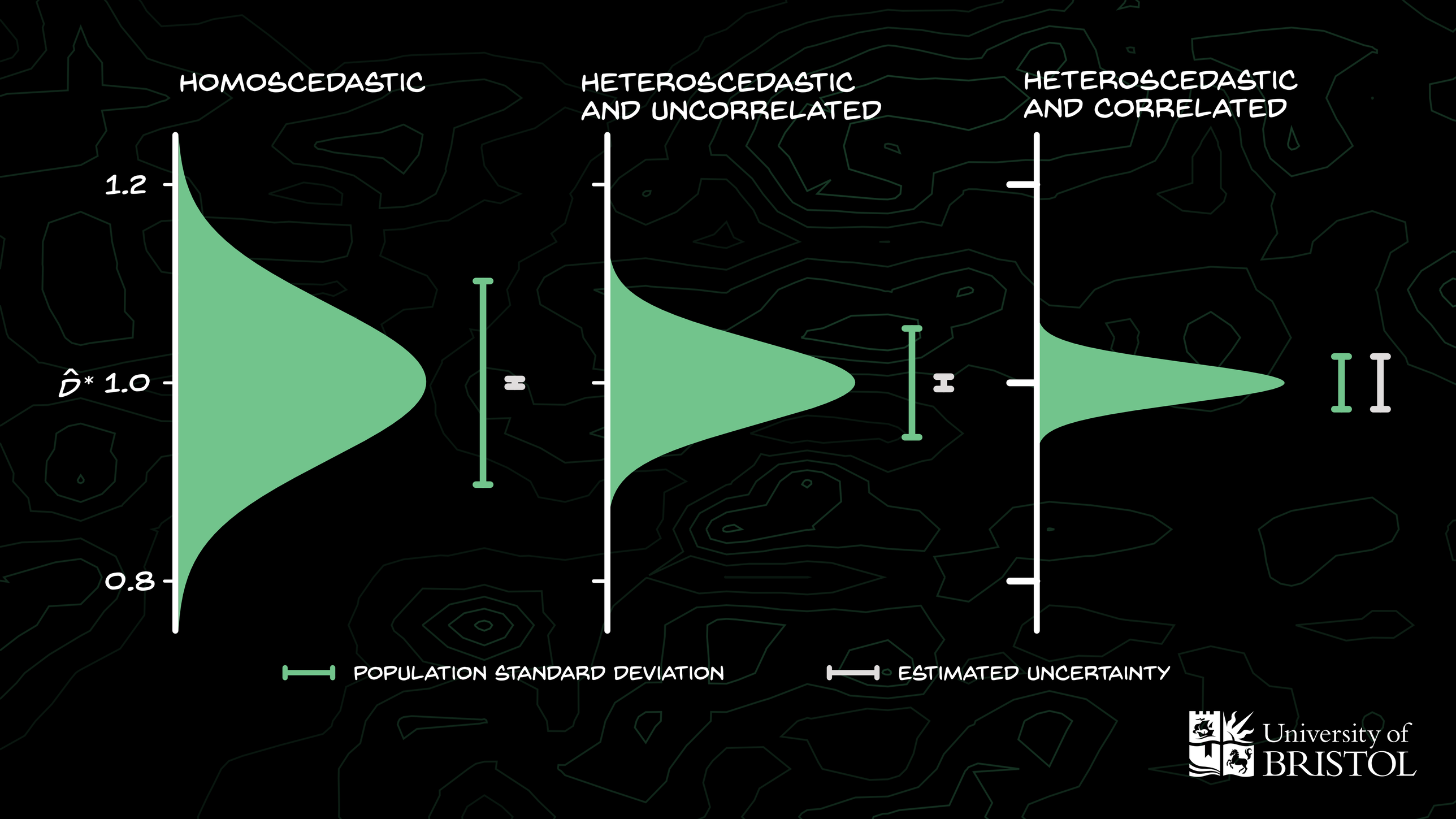
HOMOSCEDASTIC
HETEROSCEDASTIC AND UNCORRELATED
HETEROSCEDASTIC AND CORRELATED
1.2
$\hat{D}^*$ 1.0
0.8
POPULATION STANDARD DEVIATION
ESTIMATED UNCERTAINTY
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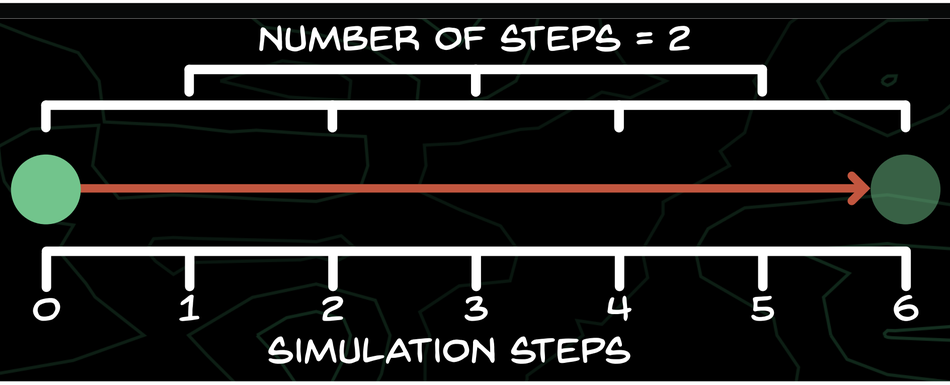
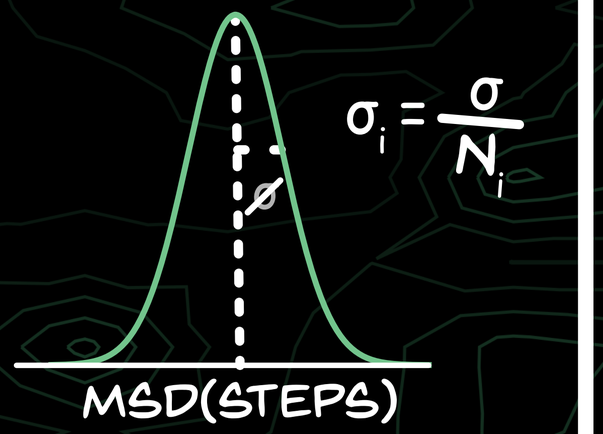
OVERLAPPING SAMPLES CAN MAXIMISE THE NUMBER OF OBSERVATIONS...



...HOWEVER, AS THE SAMPLES ARE ***NOT*** *STATISTICALLY INDEPENDENT,* THE VARIANCE MUST BE RESCALED BY THE NUMBER OF INDEPENDENT SAMPLES.



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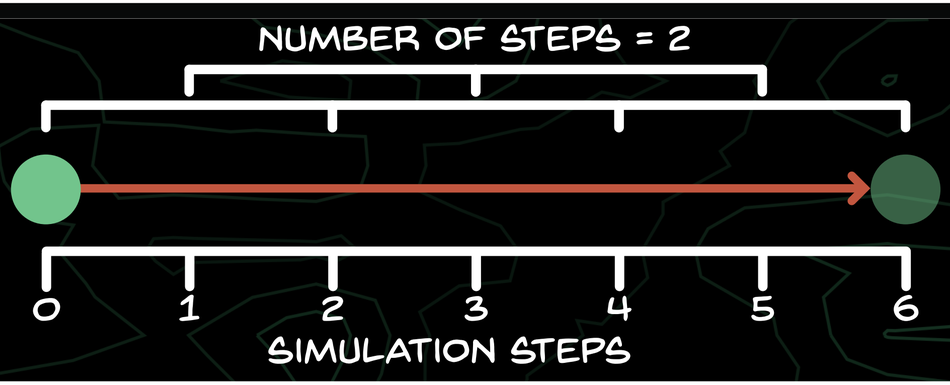

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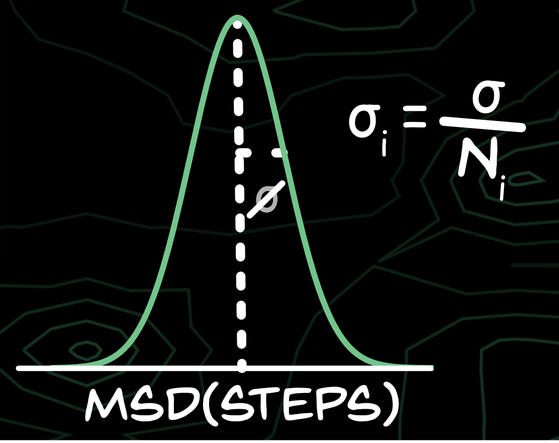
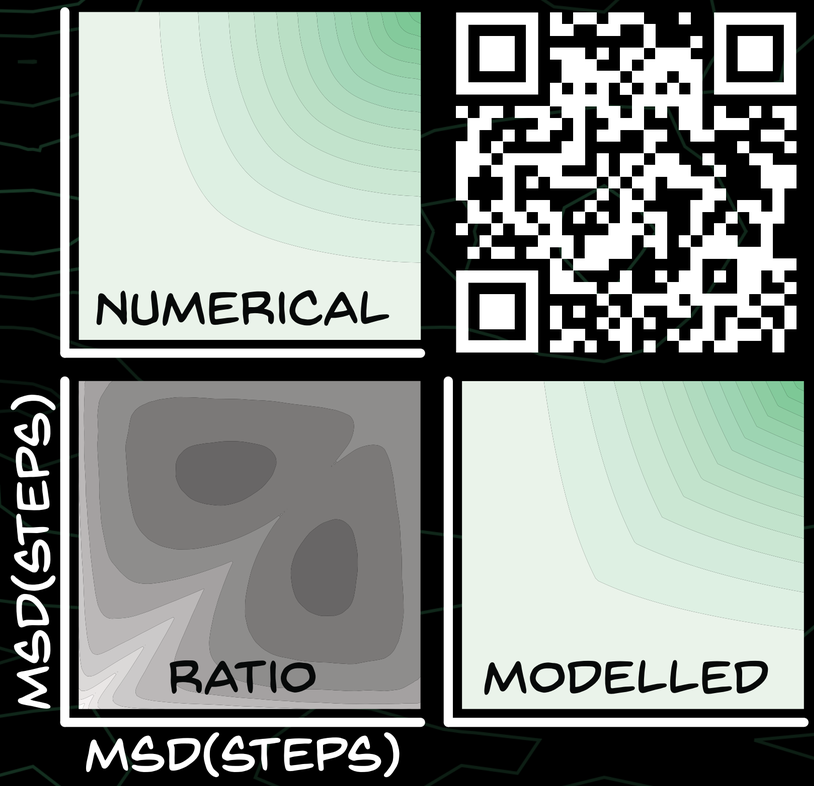


THE FULL COVARIANCE MATRIX CAN BE MODELLED BASED ON FREELY DIFFUSING PARTICLES.



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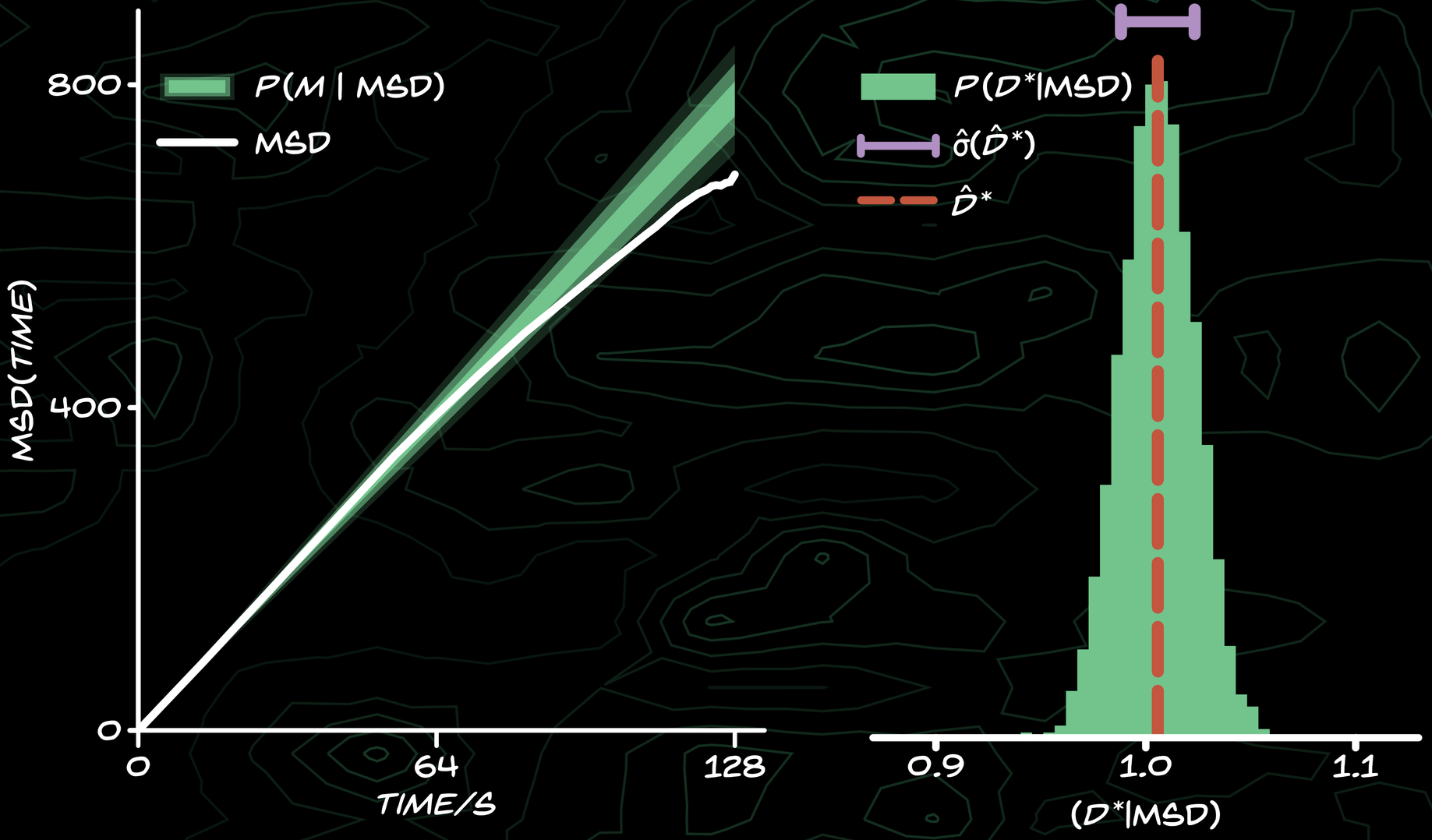

McCluskey, Coles, Morgan, *J. Chem. Theory Comput.*, **21**(1), 79, 2025.

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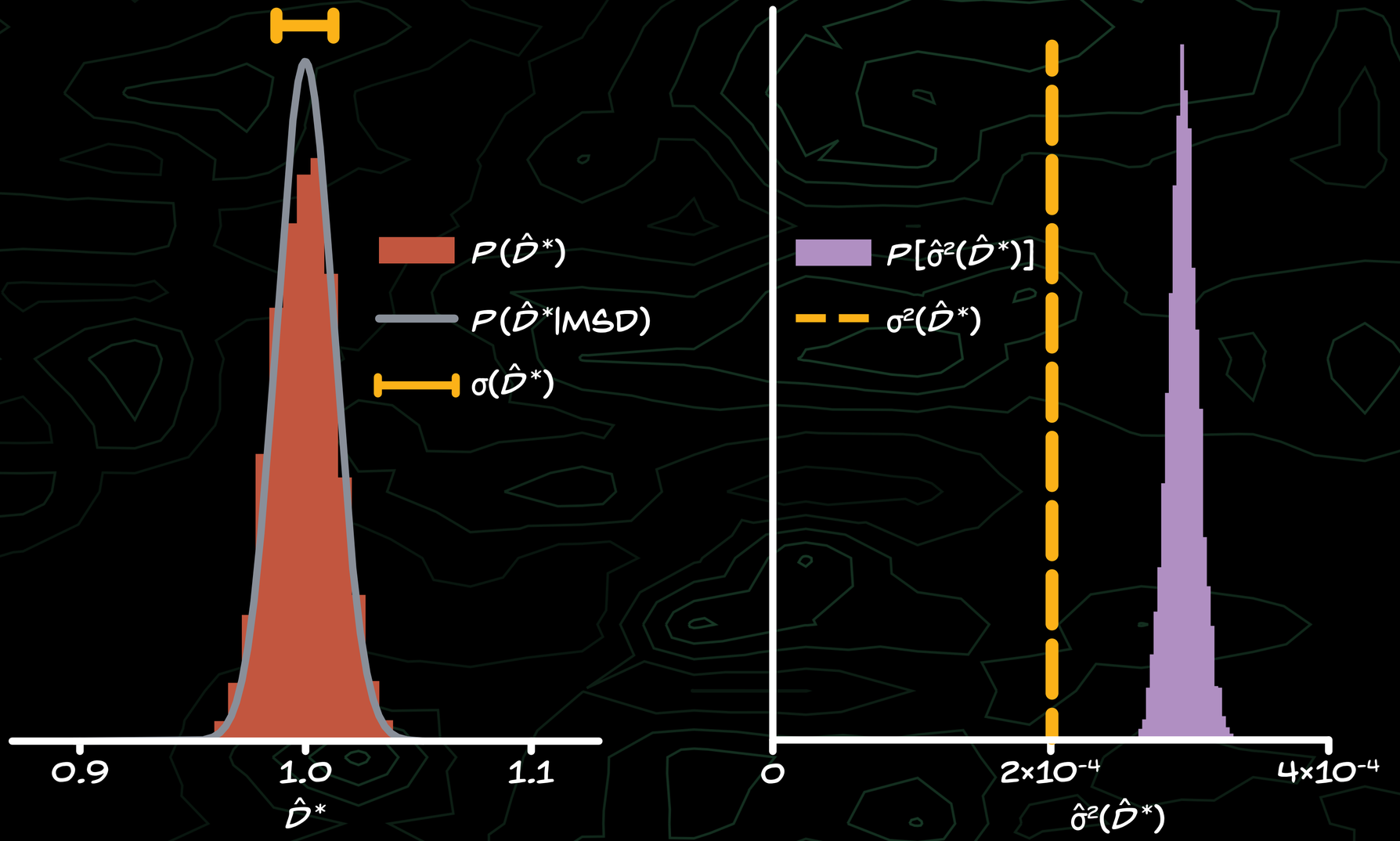

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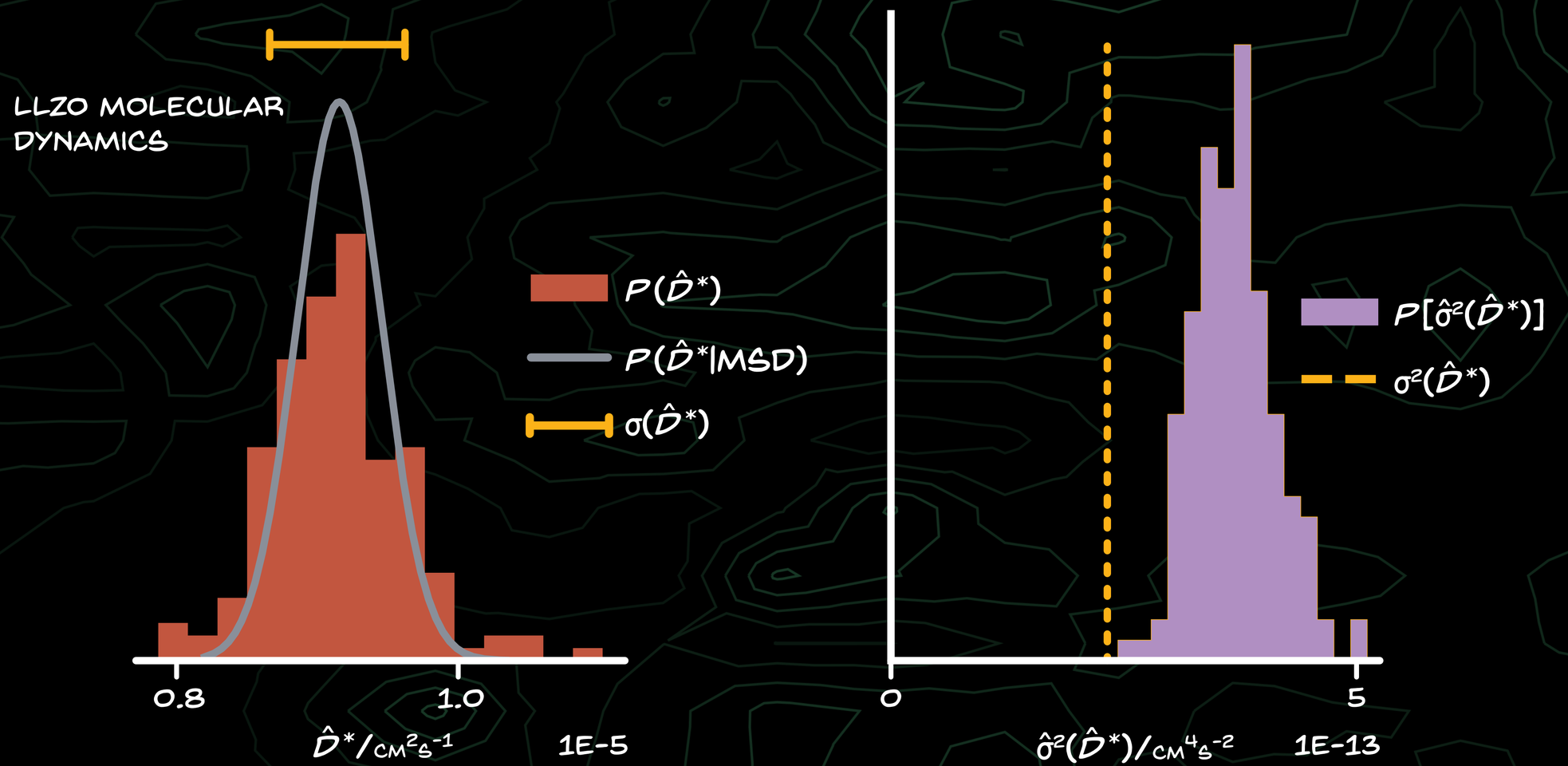

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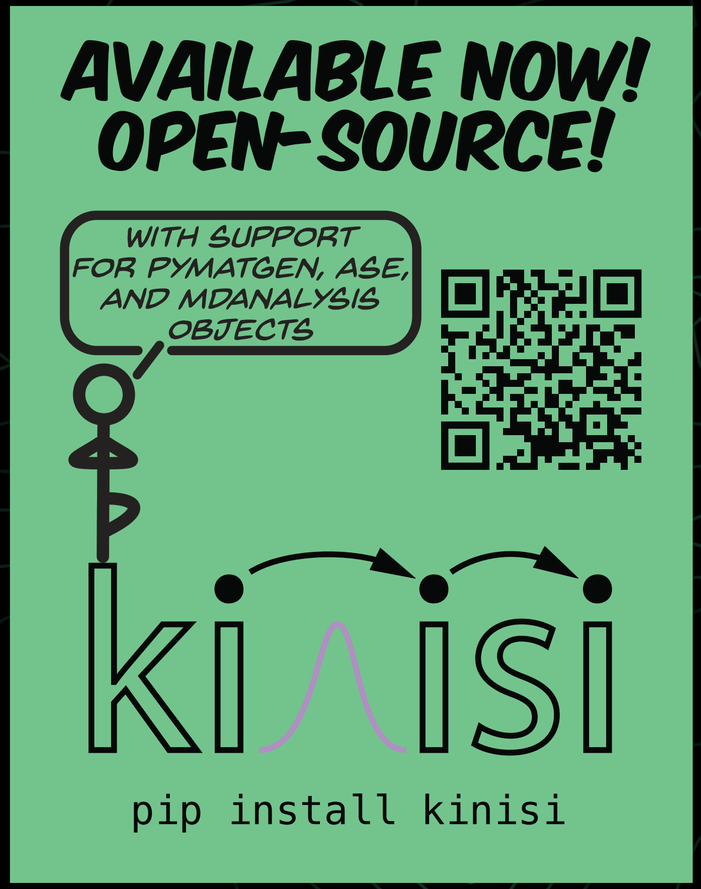

KINISI.RTFD.IO

McCluskey, Squires, Dunn, Coles, Morgan, *J. Open Source Softw.*, **9**(94), 5984, 2024.

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ARRHENIUS MODELLING CAN BE USED FOR DIFFUSION...

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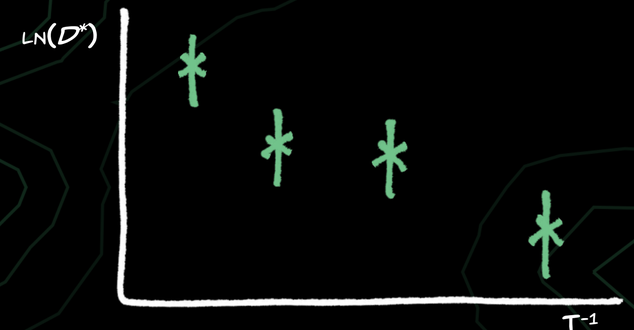

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ARRHENIUS MODELLING CAN BE USED FOR DIFFUSION...

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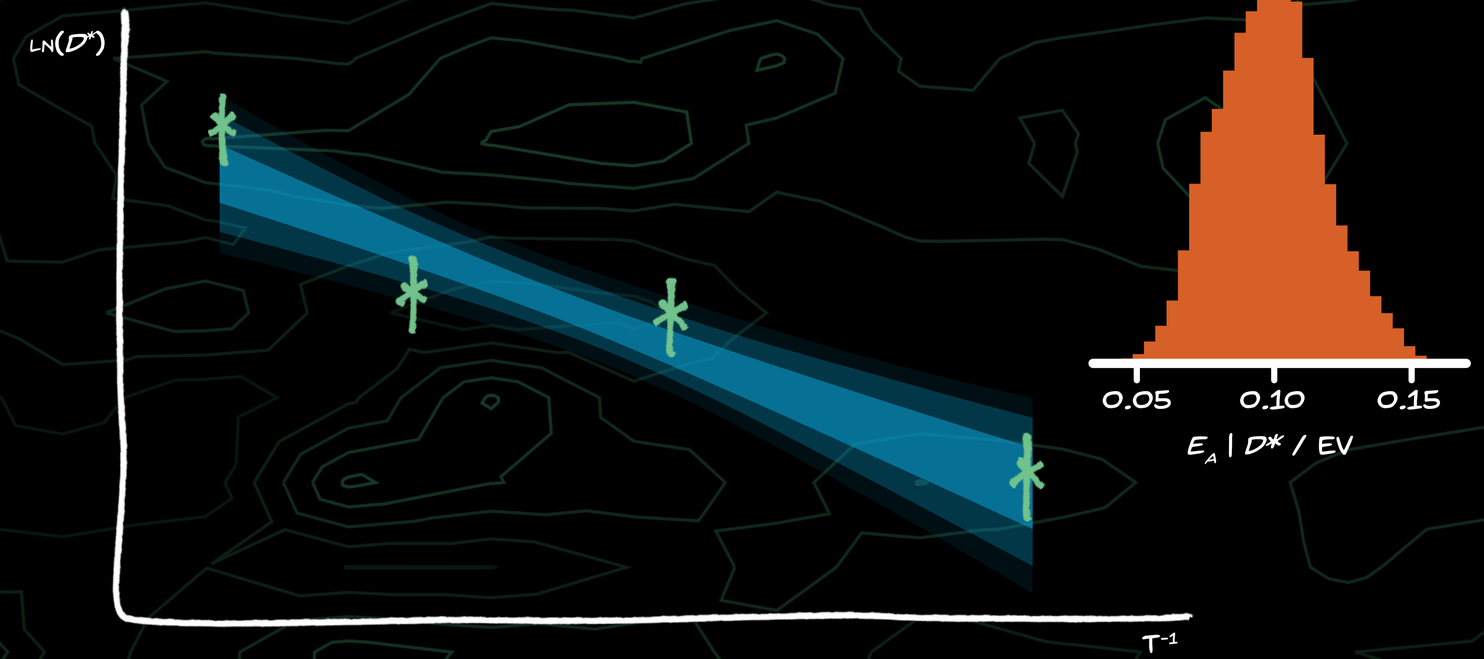
WE CAN THEN PROPAGATE THE ERROR IN *D** TO THE ERROR IN THE ACTIVATION ENERGY OF DIFFUSION.



McCluskey, *J. Chem. Educ.*, **100**(11), 4714, 2023; McCluskey, Coles, Morgan, *In Preparation.*

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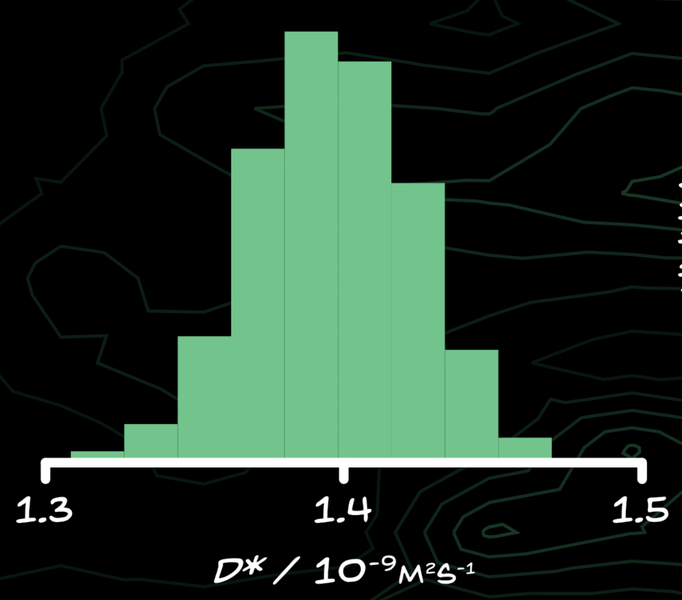
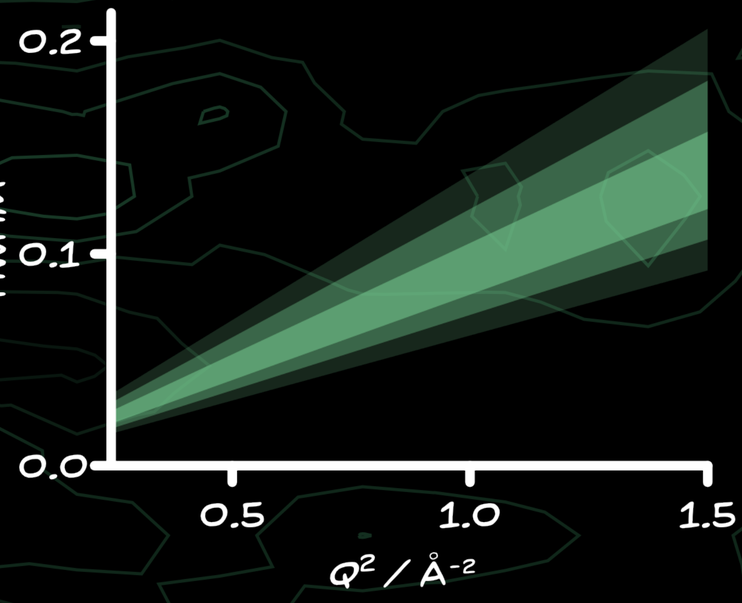
WELL BUSTER, WE CAN USE PRIOR KNOWLEDGE FROM SIMULATION IN OUR QENS ANALYSIS.*





*THE PHD PROJECT OF HARRY RICHARDSON, WHO UNFORTUNATELY COULD NOT ATTEND.

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ATOMS, IONS, OR MOLECULES MIGHT PREFER TO MOVE IN OPPOSITE DIRECTIONS...

OH GOD, NOT HIM AGAIN.

...OR PERHAPS THEY DON'T INTERACT....

I DON'T THINK ABOUT YOU AT ALL.

...OR THEY MAY SHOW COLLECTIVE CONSTRUCTIVE MOTION.

WAIT! WE ARE COMING TOO.

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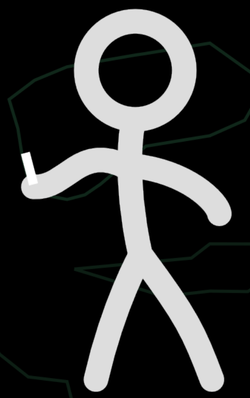
The Nernst-Einstein Relation

albert woz here

$$\sigma = \frac{Cq^2}{k_b T} D^*$$



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The Nernst-Einstein Relation

albert woz here

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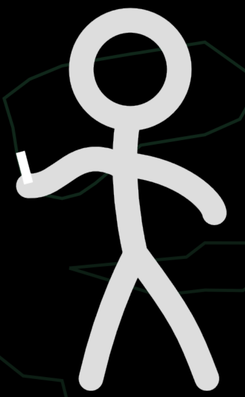

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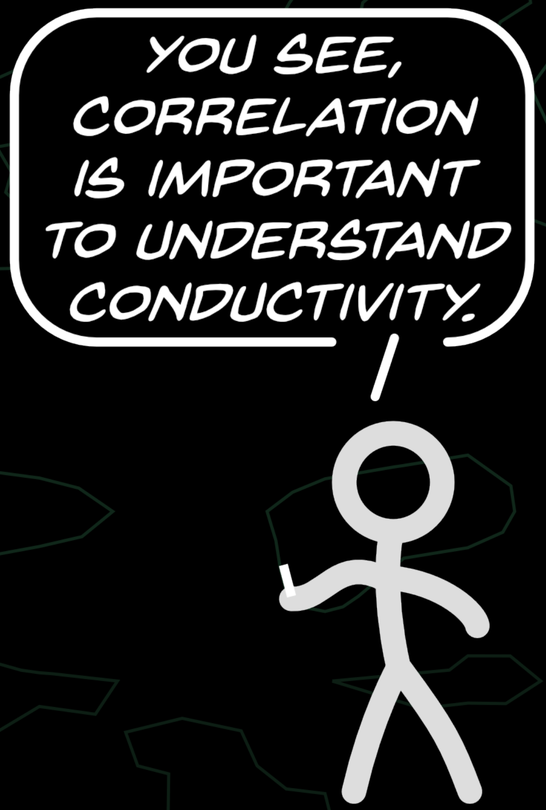
The Nernst-Einstein Relation

$$\sigma = \frac{Cq^2}{k_b T} \frac{D^*}{H_R}$$

albert woz here

~~$\sigma = \frac{Cq^2}{k_b T} D^*$~~

$$H_R = \frac{D^*}{D_\sigma}$$



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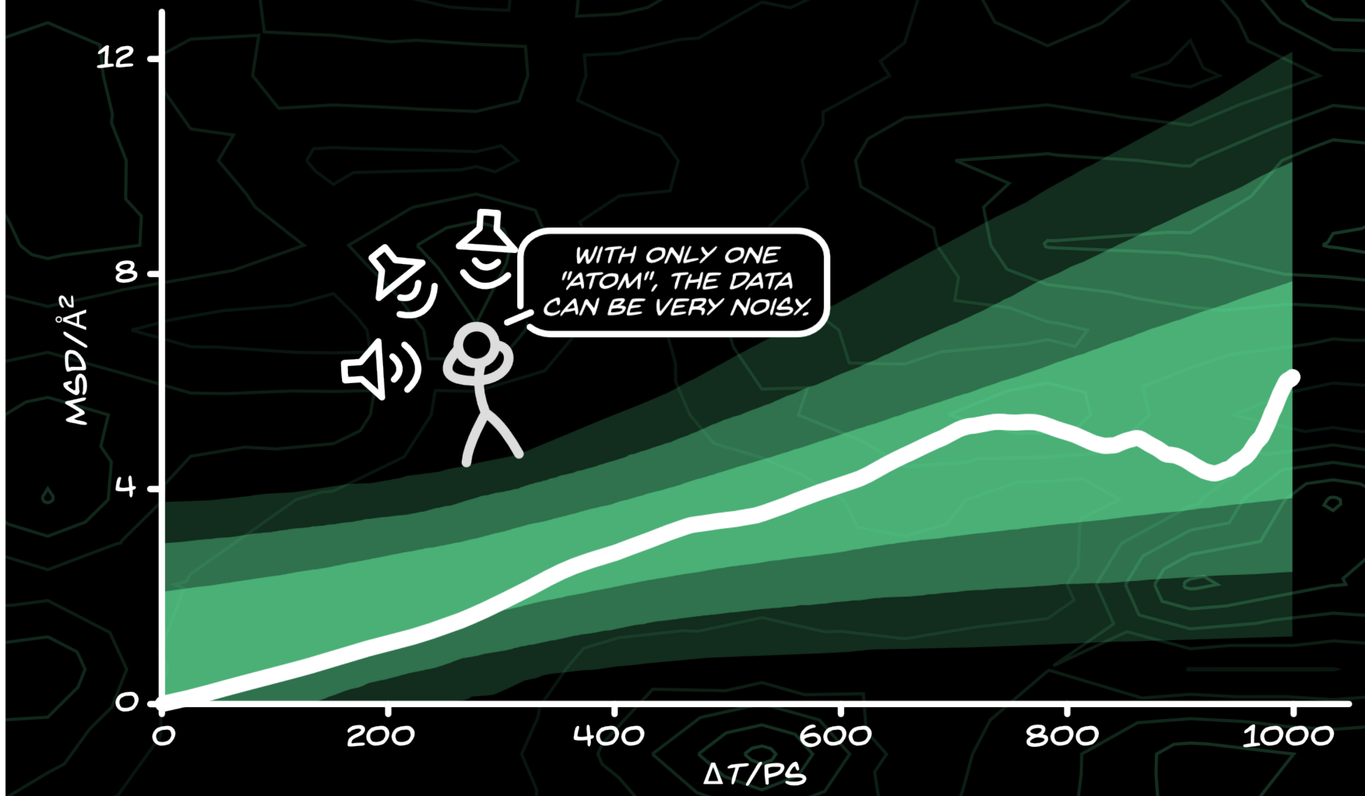
TO COMPUTE D_σ AND, THEREFORE H_R, WE NEED TO COMPUTE THE CENTRE-OF MASS MEAN-SQUARED DISPLACEMENT.



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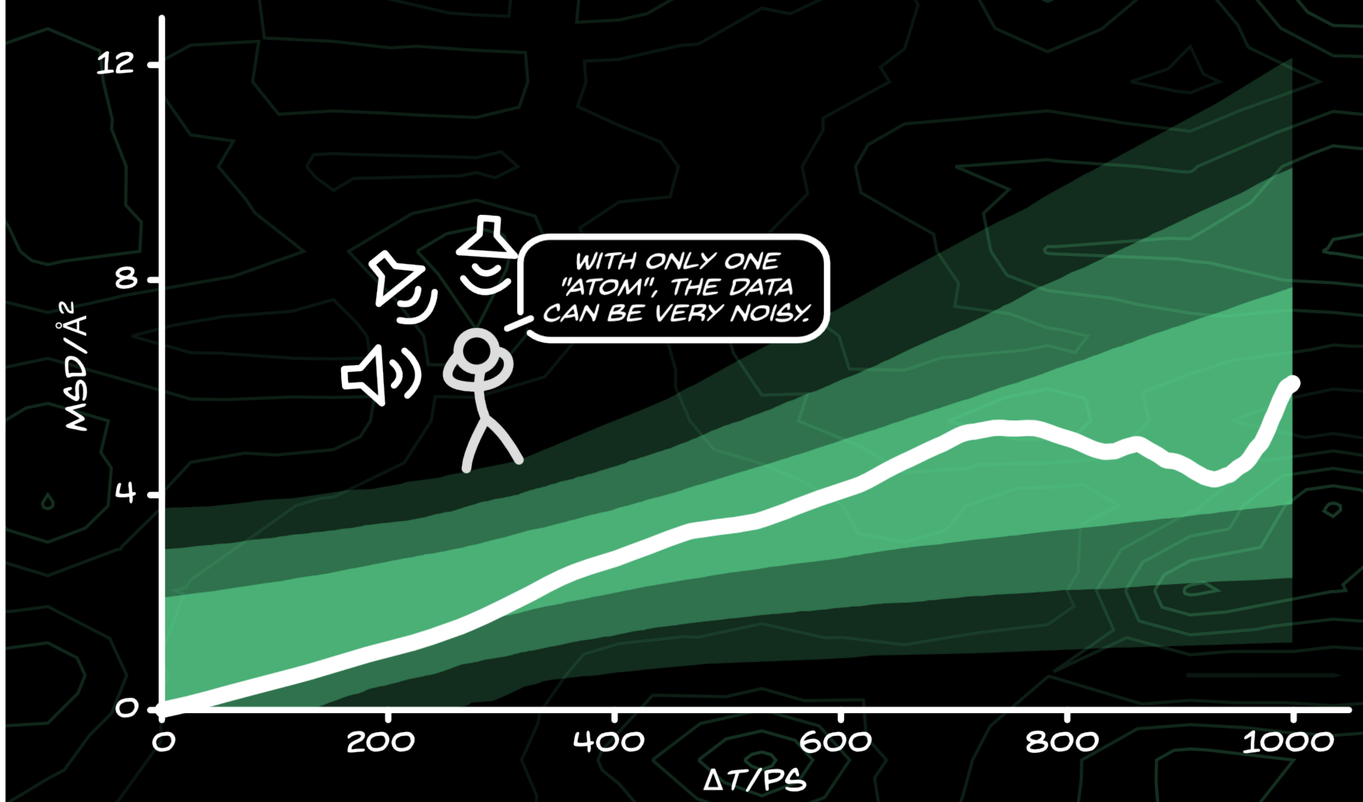
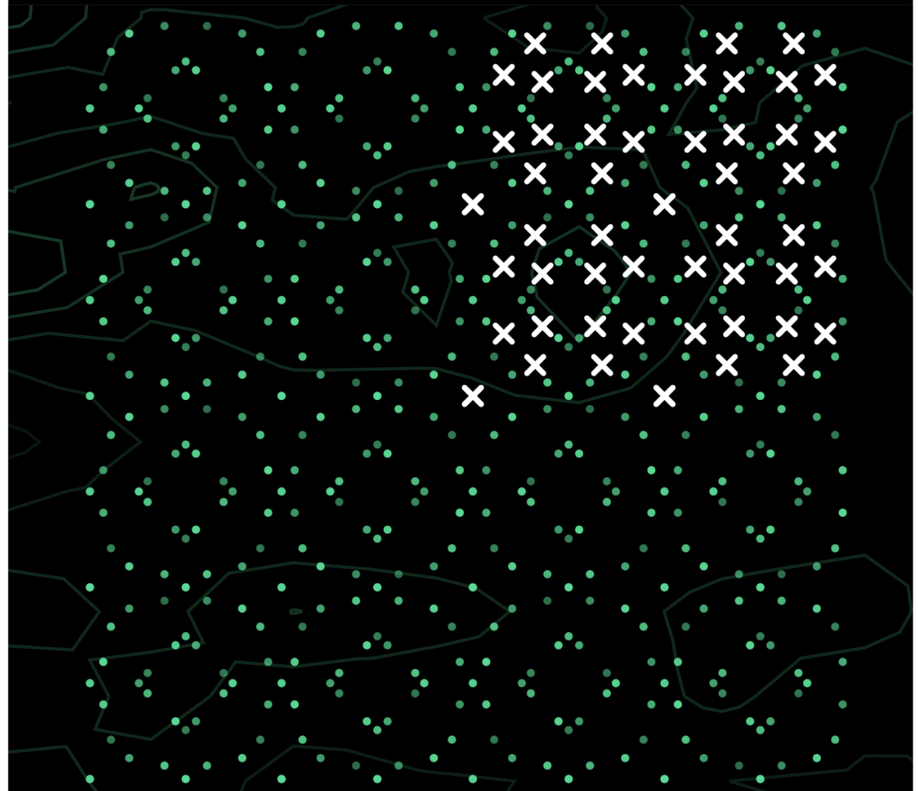


IF WE HAVE A BIG ENOUGH SIMULATION DATA, WE CAN DEFINE SUB CENTRES-OF-MASS.



2 X 2 X 2 SUPERCELL LLZO (448 LITHIUM IONS)

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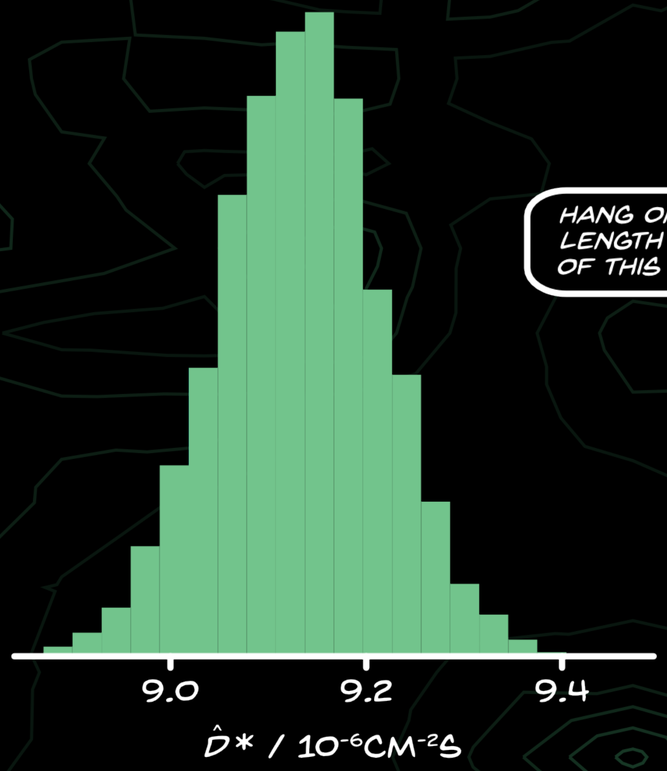
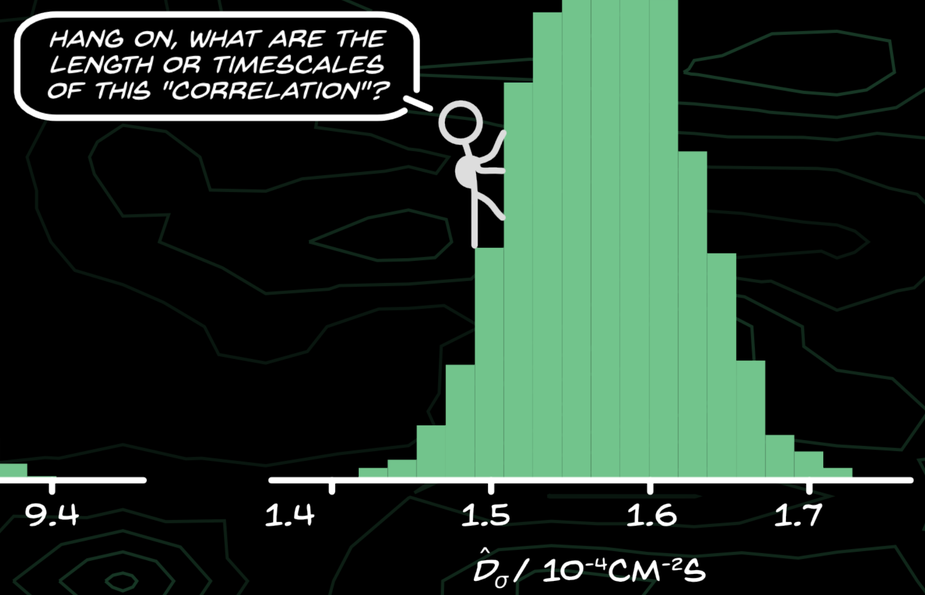
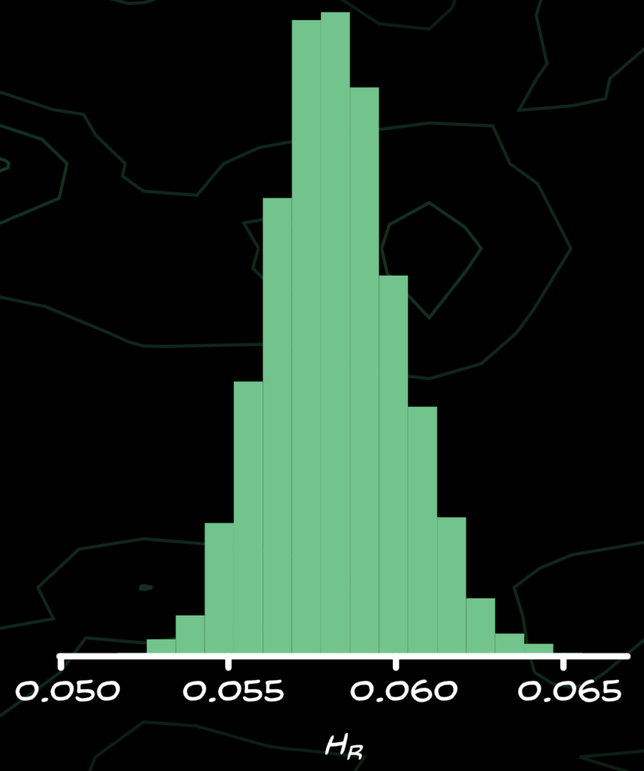
WITH THIS APPROACH, WE CAN ESTIMATE THE HAVEN RATIO, AND, THEREFORE, THE AMOUNT OF CORRELATION IN OUR SYSTEM. IT LOOKS LIKE LLZO IS HIGHLY CORRELATED?*







*THIS IS VERY NEW DATA/CODE.

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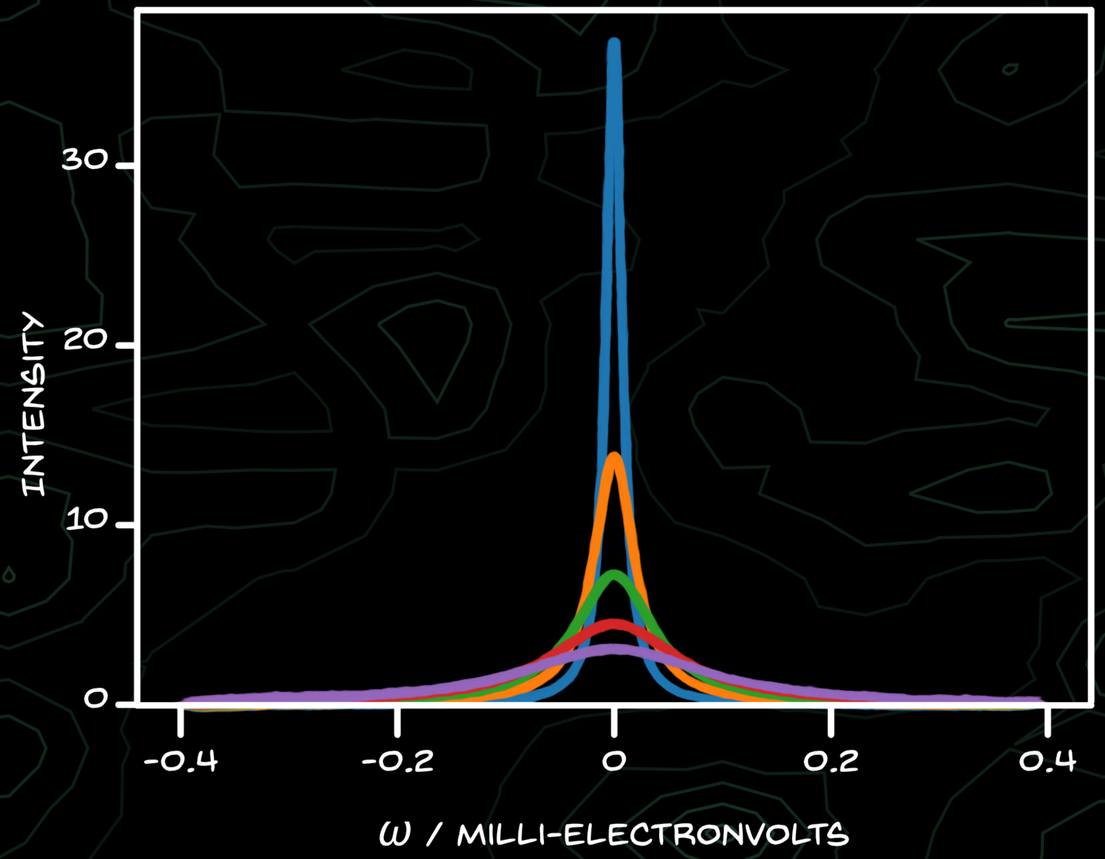
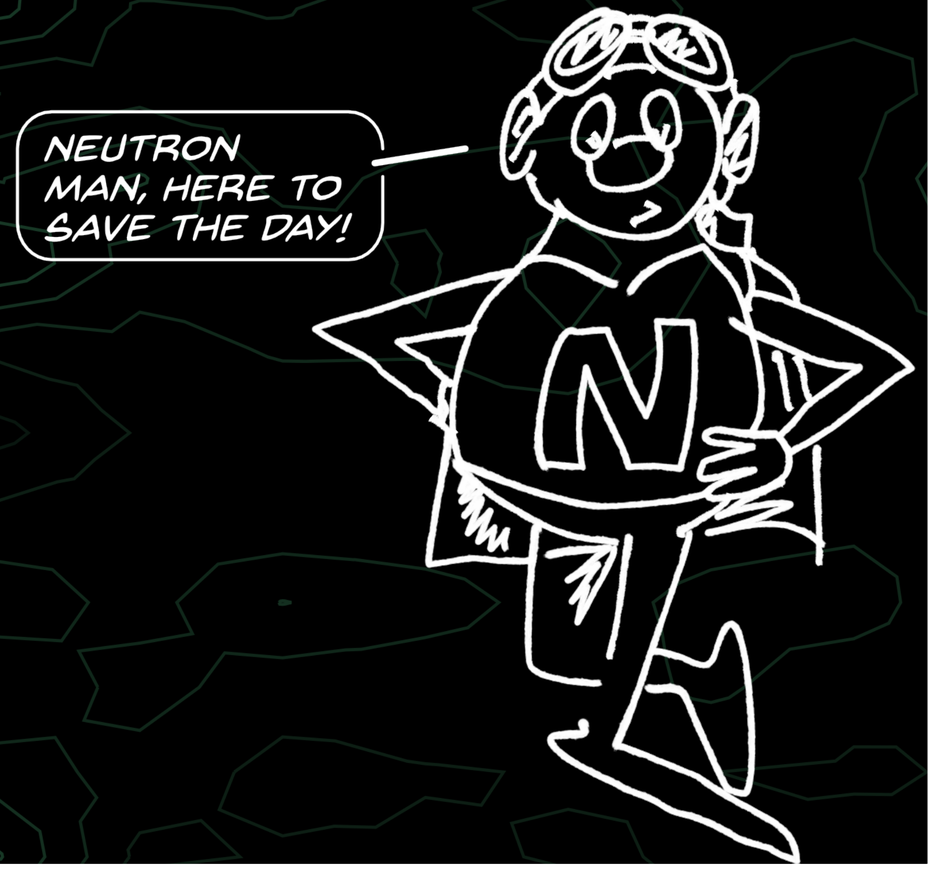
QENS IS AN EXCELLENT TOOL TO PROBE DIFFUSION...



...AND POLARISED-QENS PROVIDES AN TOOL TO MEASURE THIS CORRELATION.



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Ben Morgan (Bath)

Alexander Squires (Birmingham)

Samuel Coles (Bath/Cambridge)

Josh Dunn (Bristol)

Harry Richardson (Bristol)

Jeff Armstrong (ISIS)

Kit McColl (Bath)

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