# Universality class of the phase transition in dipolar linear rotor chains

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# Chemical species on a lattice

- peapods: $C_{60}$@CNTs[1]
- endofullerenes: $H_2@C_{60}$[2], HF@$C_{60}$[3]
- endofullerene peapods: $Dy_3N@C_{80}$@CNTs[4], $Er_3N@C_{80}$@CNTs[5]
- Why not HF@$C_{60}$@CNTs?
  - **Compute low-temperature properties of polar molecules in endofullerene peapods.**

[1] B. W. Smith et al., "Encapsulated $C_{60}$ in carbon nanotubes", Nature **396**, 323 (1998).

[2] K. Komatsu et al., "Encapsulation of molecular hydrogen in fullerene $C_{60}$ by organic synthesis", Science **307**, 238–240 (2005).

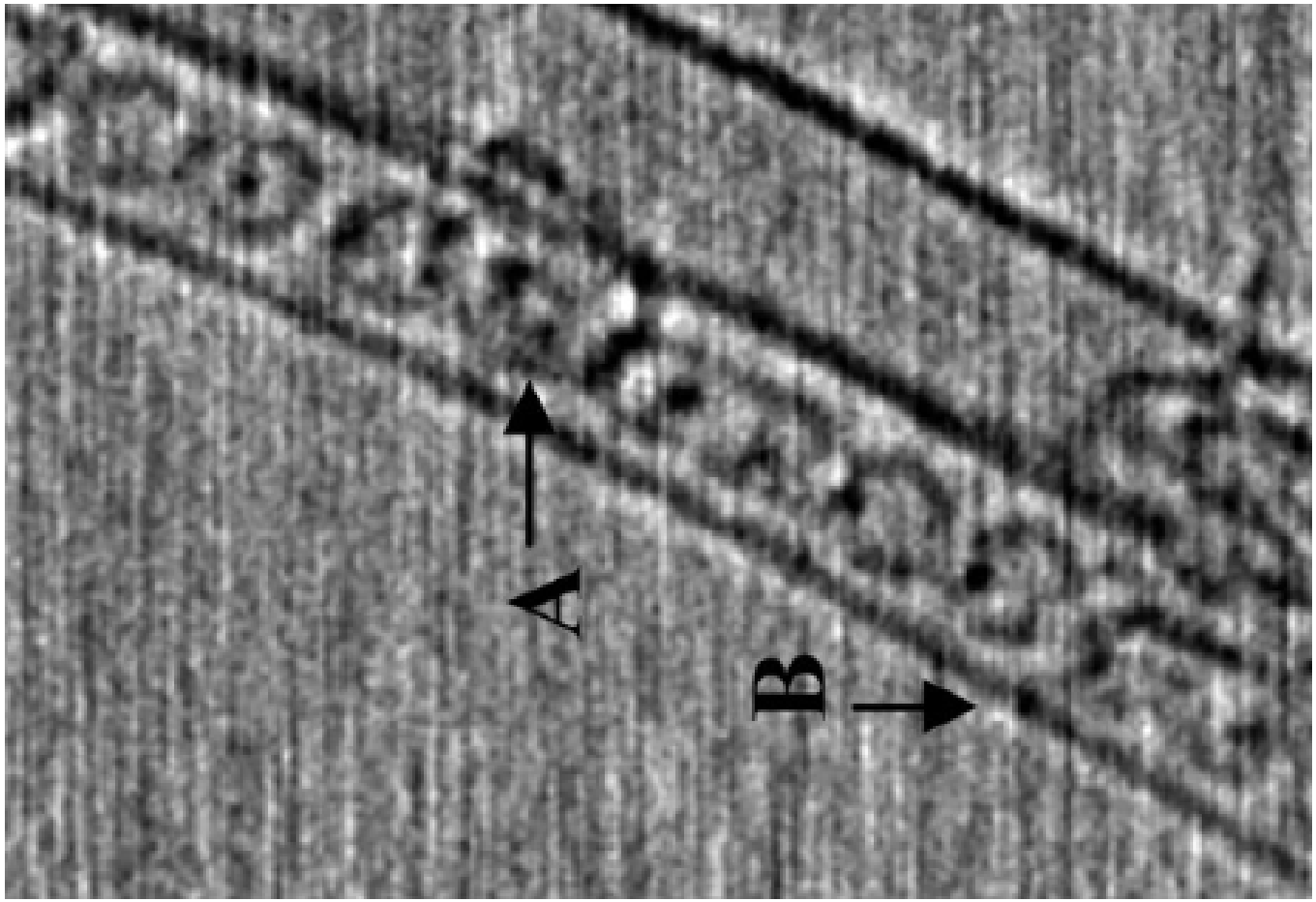
[3] A. Krachmalnicoff et al., "The dipolar endofullerene HF@$C_{60}$", Nature Chemistry **8**, 953–957 (2016).

[4] H. Shiozawa et al., "Filling factor and electronic structure of $Dy_3N@C_{80}$ filled single-wall carbon nanotubes studied by photoemission spectroscopy", Phys. Rev. B **73**, 205411 (2006).

[5] F. Fritz et al., "Nanoscale x-ray investigation of magnetic metallofullerene peapods", Nanotechnology **28**, 435703 (2017).

# Chemical species on a lattice

## $La@C_{82}$ fullerene peapods:




[1] R. J. Nicholls et al., "Direct imaging and chemical identification of the encapsulated metal atoms in bimetallic endofullerene peapods", ACS Nano 4, 3943–3948 (2010)

# Simplified model

- Too many degrees of freedom!
- Full analysis includes:
  - translation, rotation, vibration of each molecule
  - electrostatic interactions between molecules
  - interactions with fullerenes, nanotube

- Simplifying assumptions:
  - motion of molecule inside cage is small and slow
  - molecules are linear and rigid
  - dipolar interactions are dominant

# Simplified model

- dipolar linear rotor lattice along $z$:

$$\hat{H} = \frac{B}{\hbar^2} \sum_{i=1}^{N} \hat{\ell}_i^2 + \frac{\mu^2}{4\pi\epsilon_0 R^3} \sum_{i<j} \frac{\hat{\mathbf{e}}_i \cdot \hat{\mathbf{e}}_j - 3\hat{z}_i \hat{z}_j}{|i-j|^3}$$

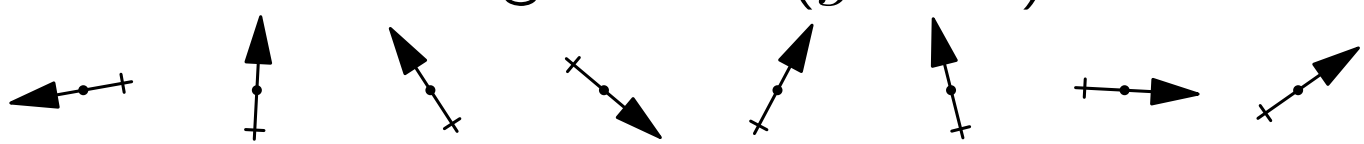
- removed most of the chemistry from the model, but some still remains
- non-dimensionalized, single-parameter version:

$$\hat{H} = \sum_{i=1}^{N} \hat{\ell}_i^2 + g \sum_{i<j} \frac{\hat{\mathbf{e}}_i \cdot \hat{\mathbf{e}}_j - 3\hat{z}_i \hat{z}_j}{|i-j|^3}$$

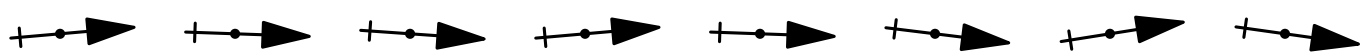
# Simplified model

$$\hat{H} = \sum_{i=1}^{N} \hat{\ell}_i^2 + g \sum_{i<j} \frac{\hat{\mathbf{e}}_i \cdot \hat{\mathbf{e}}_j - 3\hat{z}_i \hat{z}_j}{|i-j|^3}$$

- disordered configuration ($g \ll 1$):



- ordered configuration ($g \gg 1$):



# Exact diagonalization

- Construct sparse Hamiltonian, use iterative diagonalization to find ground state[1]
  - Exact diagonalization does not scale well with system size

THE JOURNAL OF CHEMICAL PHYSICS **148**, 074112 (2018)

**Quantifying entanglement of rotor chains using basis truncation: Application to dipolar endofullerene peapods**

Tom Halverson, Dmitri Iouchtchenko, and Pierre-Nicholas Roy
*Department of Chemistry, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada*

(Received 2 November 2017; accepted 5 February 2018; published online 21 February 2018)

We propose a variational approach for the calculation of the quantum entanglement entropy of assemblies of rotating dipolar molecules. A basis truncation scheme based on the total angular momentum quantum number is proposed. The method is tested on hydrogen fluoride (HF) molecules confined in

[1] T. Halverson et al., "Quantifying entanglement of rotor chains using basis truncation: Application to dipolar endofullerene peapods", Journal of Chemical Physics 148, 074112 (2018).

# Path integral Monte Carlo

- Path integral ground state (PIGS) Monte Carlo to compute ground state properties directly
  - Monte Carlo suffers from ergodicity issues

**Estimation of ground state entanglement entropy for continuous rotational degrees of freedom: a path integral *replica* trick approach**

Tapas Sahoo, Dmitri Iouchtchenko, Chris M. Herdman, and Pierre-Nicholas Roy[a)]
*Department of Chemistry, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada*

(Dated: 20 March 2019)

Rényi entropy reveals the extent to which states of individual particles in a many–body system are entangled to each other due to their non–local correlations. The main purpose of this study is to investigate the effect of interaction strength on the second Rényi entropy of a many–body system as a measure of entanglement for continuous rotational degrees of freedom. As the Rényi entropy is defined in relation to purity of states

# Density matrix renormalization group

- One-dimensional problem, so we use the density matrix renormalization group (DMRG)
- DMRG is a very powerful tool for finding ground states of strongly coupled condensed-matter Hamiltonians[1]

Density matrix formulation for quantum renormalization groups [PDF] ap

SR White - Physical review letters, 1992 - APS

A generalization of the numerical renormalization-group procedure used first by Wilson for the Kondo problem is presented. It is shown that this formulation is optimal in a certain sense. As a demonstration of the effectiveness of this approach, results from numerical real …

☆ Cited by 4969 Related articles »

[1] S. R. White, "Density matrix formulation for quantum renormalization groups", Phys. Rev. Lett. **69**, 2863 (1992).

# Density matrix renormalization group

- Scales easily to 100 rotors[1]
  - Rotors have infinite basis of spherical harmonics $|\ell\, m\rangle$, which we truncate at $\ell_{\max}$

THE JOURNAL OF CHEMICAL PHYSICS **148**, 134115 (2018)

**Ground states of linear rotor chains via the density matrix renormalization group**

Dmitri Iouchtchenko and Pierre-Nicholas Roy[a)]
*Department of Chemistry, University of Waterloo, Waterloo, Ontario N2L 3G1, Canada*

(Received 31 January 2018; accepted 26 March 2018; published online 4 April 2018)

In recent years, experimental techniques have enabled the creation of ultracold optical lattices of molecules and endofullerene peapod nanomolecular assemblies. It was previously suggested that the rotor model resulting from the placement of dipolar linear rotors in one-dimensional lattices at low temperature has a transition between ordered and disordered phases. We use the density matrix renormalization group (DMRG) to compute ground states of chains of up to 100 rotors and provide further

[1] D. Iouchtchenko and P.-N. Roy, "Ground states of linear rotor chains via the density matrix renormalization group", Journal of Chemical Physics 148, 134115 (2018).

# Von Neumann entropy

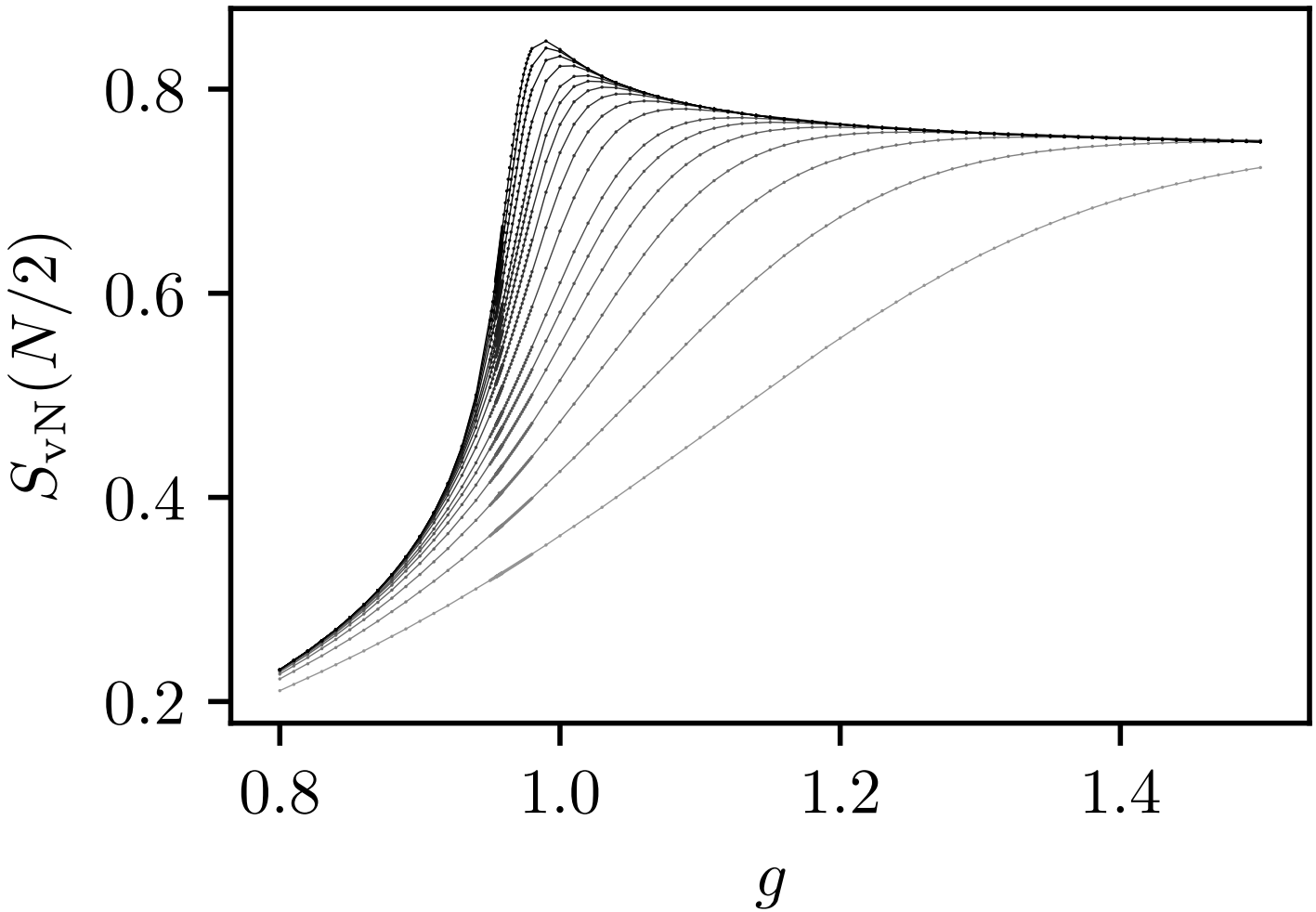
- Schmidt decomposition:

$$|\psi_{AB}\rangle = \sum_{k=1}^{M} \sqrt{\lambda_k}\, |k_A\rangle\, |k_B\rangle$$

- for a normalized state $|\psi_{AB}\rangle$, $\sum_k \lambda_k = 1$
- because $0 \leq \lambda_k \leq 1$, we can interpret this as a probability distribution
- Shannon entropy: $-\sum_{k=1}^{M} \lambda_k \log \lambda_k$
- von Neumann entropy: $S_{\mathrm{vN}} = -\sum_{k=1}^{M} \lambda_k \log \lambda_k$
- von Neumann entropy: $S_{\mathrm{vN}} = -\operatorname{Tr} \varrho_A \log \varrho_A$
  - $\varrho_A = \operatorname{Tr}_B |\psi_{AB}\rangle\langle\psi_{AB}|$

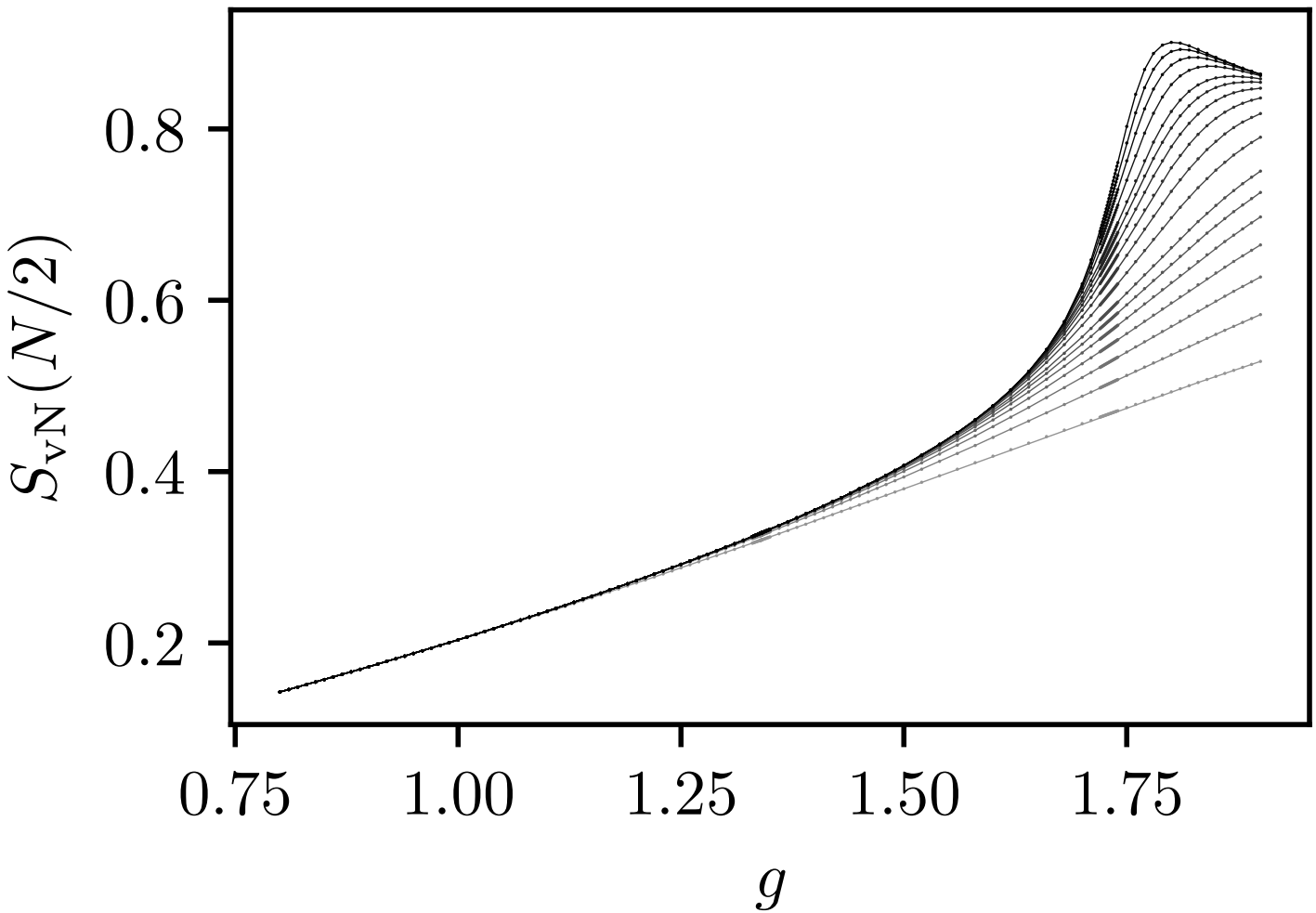
# Von Neumann entropy

- All interactions, $\ell_{\max} = 3$, $N = 8$ to $144$



# Von Neumann entropy

- Nearest-neighbour interactions, $\ell_{\max} = 1$, $N = 8$ to $144$



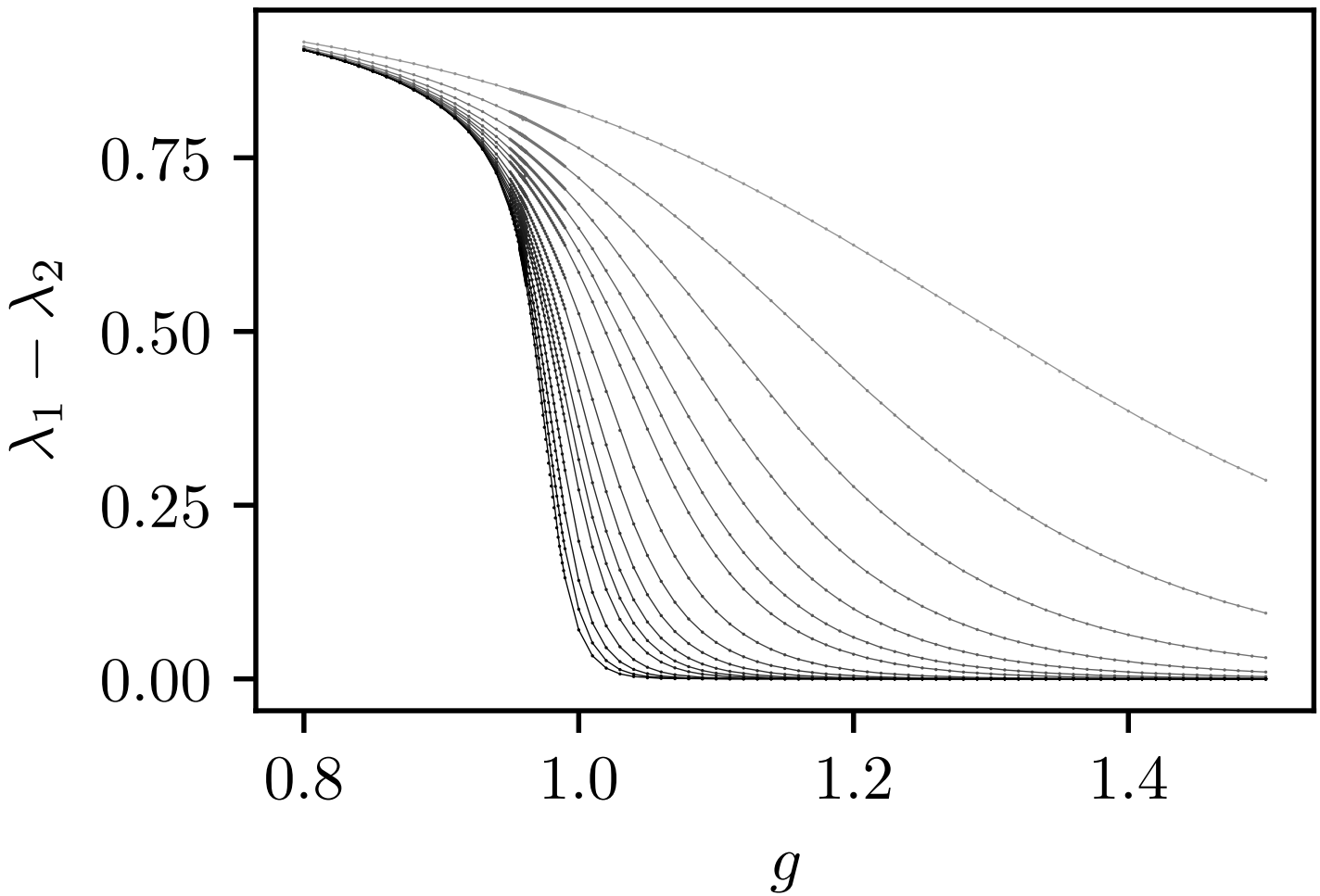
# Other properties

- Schmidt gap:[1] $\lambda_1 - \lambda_2$
  - Vanishes when dominant Schmidt coefficients become degenerate
- Energy gap: $E_1 - E_0$
  - Vanishes when timescales diverge
- Orientational correlation: $\frac{2}{N(N-1)} \left\langle \sum_{i<j} \hat{\mathbf{e}}_i \cdot \hat{\mathbf{e}}_j \right\rangle$
  - Vanishes in disordered states

[1] G. De Chiara et al., "Entanglement spectrum, critical exponents, and order parameters in quantum spin chains", Physical Review Letters **109**, 237208 (2012).

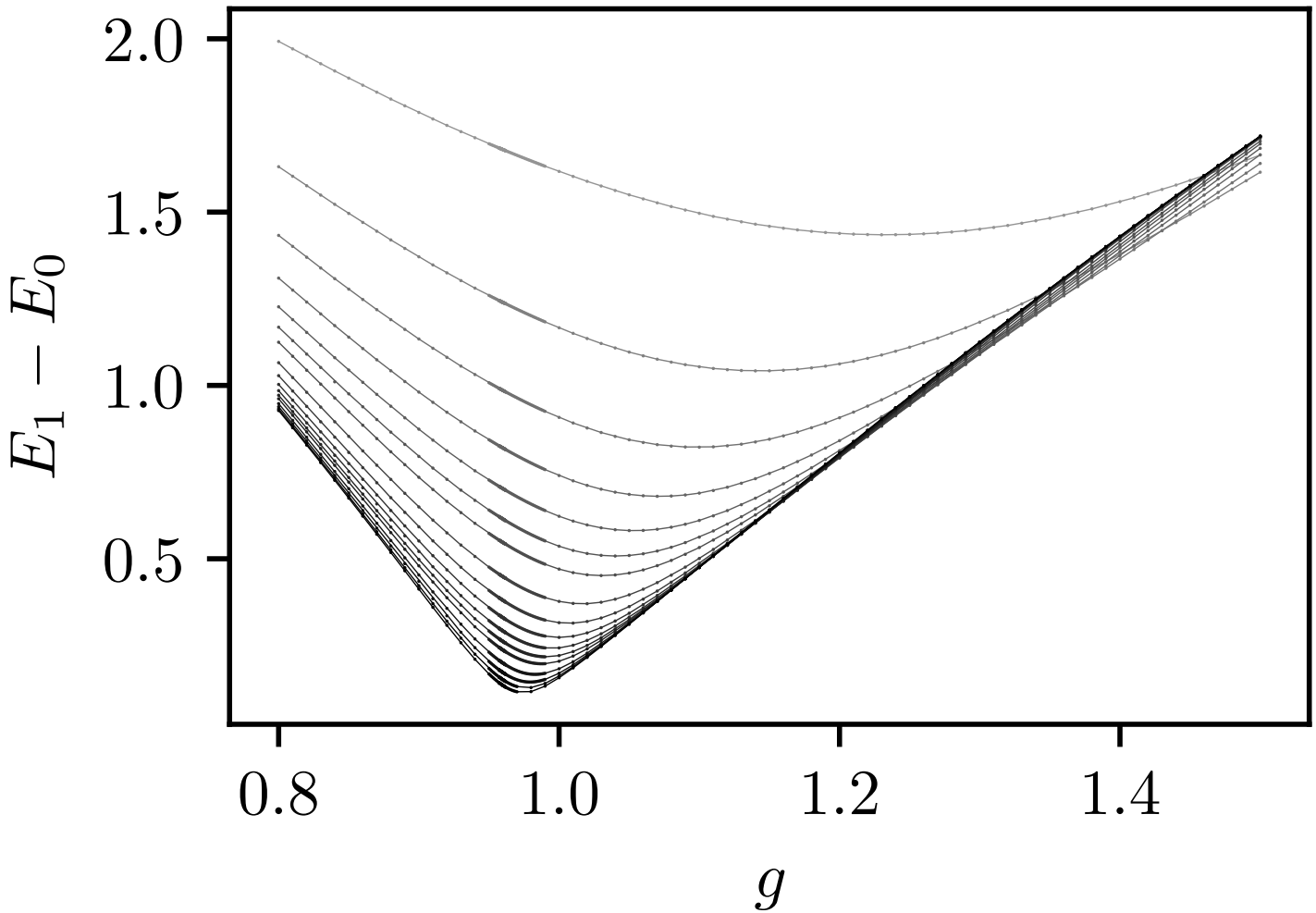
# Schmidt gap

- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



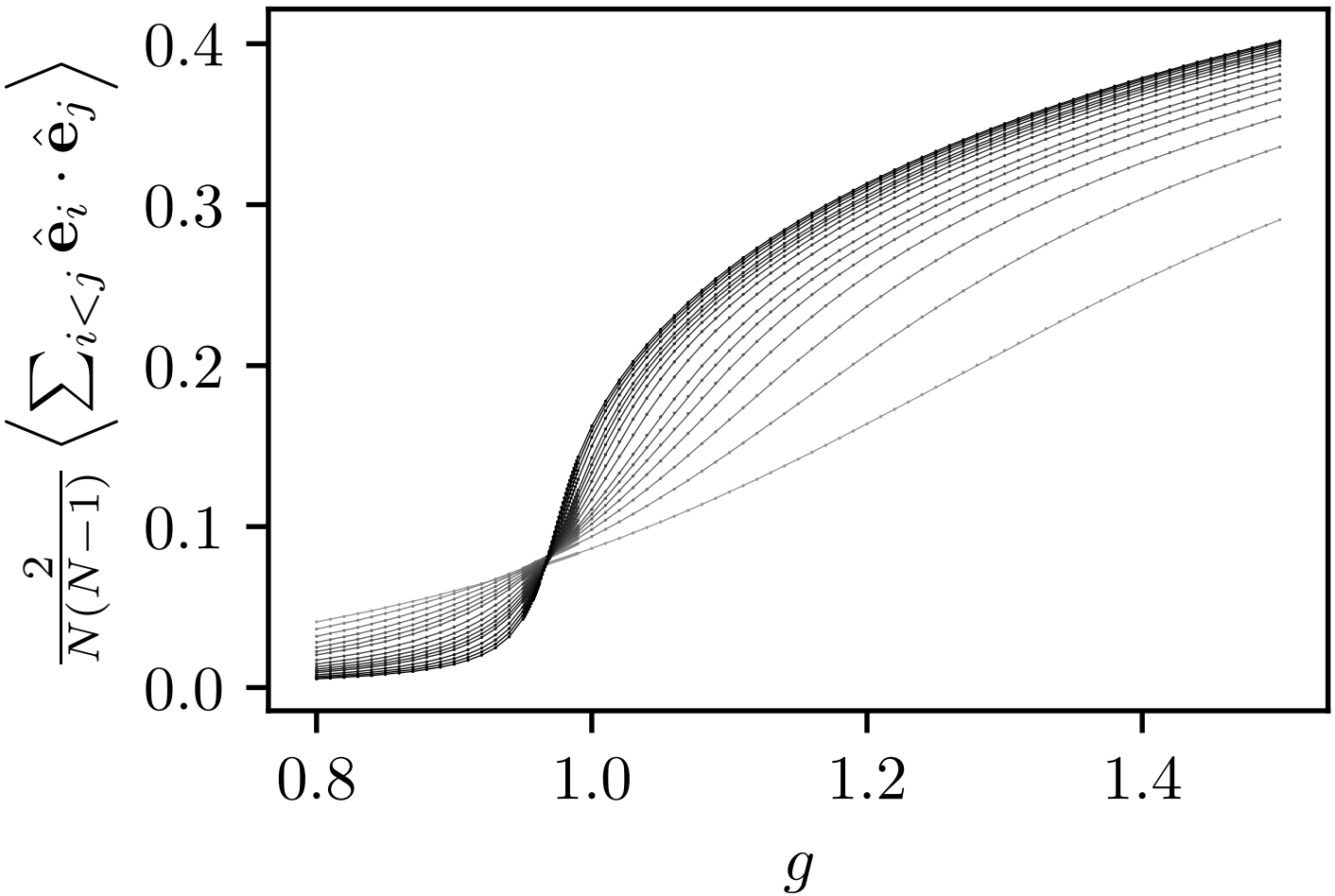
# Energy gap

- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



# Orientational correlation

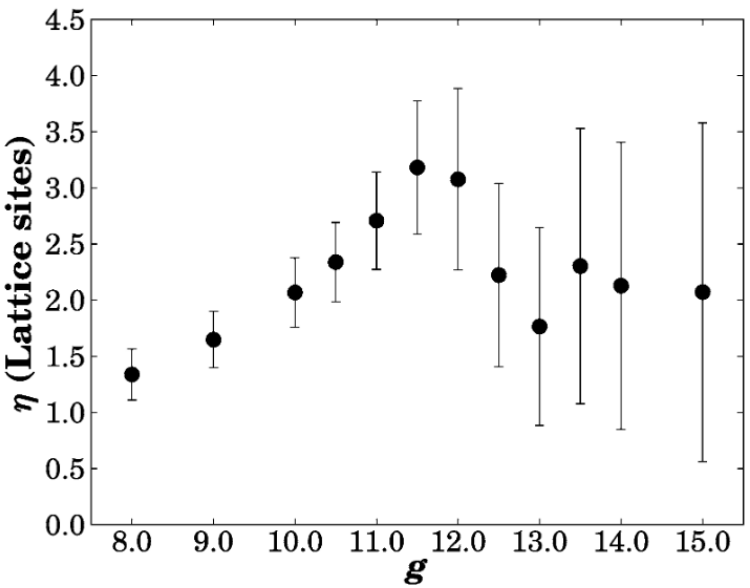
- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



# Quantum phase transition

- 2011 PIGS Monte Carlo study by Abolins et al.[1]
  - evidence of a second-order transition between ordered and disordered phases:

**Fig. 3** Correlation length $\eta$ extracted from exponential decay of $C(r)$, plotted as a function of interaction strength, $g$, for a system of 64 dipoles. Despite the increasing error bars for large $g$, a peak near $g = 11.5$ is clearly visible, signaling the phase transition from a disordered phase at low $g$ and an ordered phase at high $g$




[1] B. Abolins et al., "A ground state Monte Carlo approach for studies of dipolar systems with rotational degrees of freedom", Journal of Low Temperature Physics **165**, 249–260 (2011).

# Continuous phase transition

- In a continuous (second-order) phase transition at $g_c$, various quantities diverge[1]
  - correlation length: $\xi \sim |g - g_c|^{-\nu}$
  - energy gap: $\Delta E \sim \xi^{-z} \sim |g - g_c|^{z\nu}$
  - Schmidt gap: $\Delta\lambda \sim |g - g_c|^{\beta}$
  - susceptibility: $\chi \sim |g - g_c|^{-\gamma}$

- Critical exponents are universal!
  - depend on symmetry, dimensionality
  - independent of microscopic details

[1] S. Sachdev, **Quantum phase transitions**, 2nd ed. (Cambridge University Press, 2011).

# Scaling hypothesis

- Phase transitions are in the thermodynamic limit
- When $g \to g_c$ and $N \to \infty$, a property $K(g) \sim |g - g_c|^{-\kappa}$ has a universal scaling form:[1]

$$K(g, N) = N^{\frac{\kappa}{\nu}} \tilde{K}\left(N^{\frac{1}{\nu}}(g - g_c)\right)$$

- Data collapse:
  - $y(g, N) = N^{-\frac{\kappa}{\nu}} K(g, N)$ vs $g$:

$$y(g_c, N) = \tilde{K}(0)$$

  - $y(x, N) = N^{-\frac{\kappa}{\nu}} K(g(x), N)$ vs $x = N^{\frac{1}{\nu}}(g - g_c)$:

$$y(x, N) = \tilde{K}(x)$$

[1] R. Samajdar et al., "Numerical study of the chiral $\mathbb{Z}_3$ quantum phase transition in one spatial dimension", Physical Review A **98**, 023614 (2018).

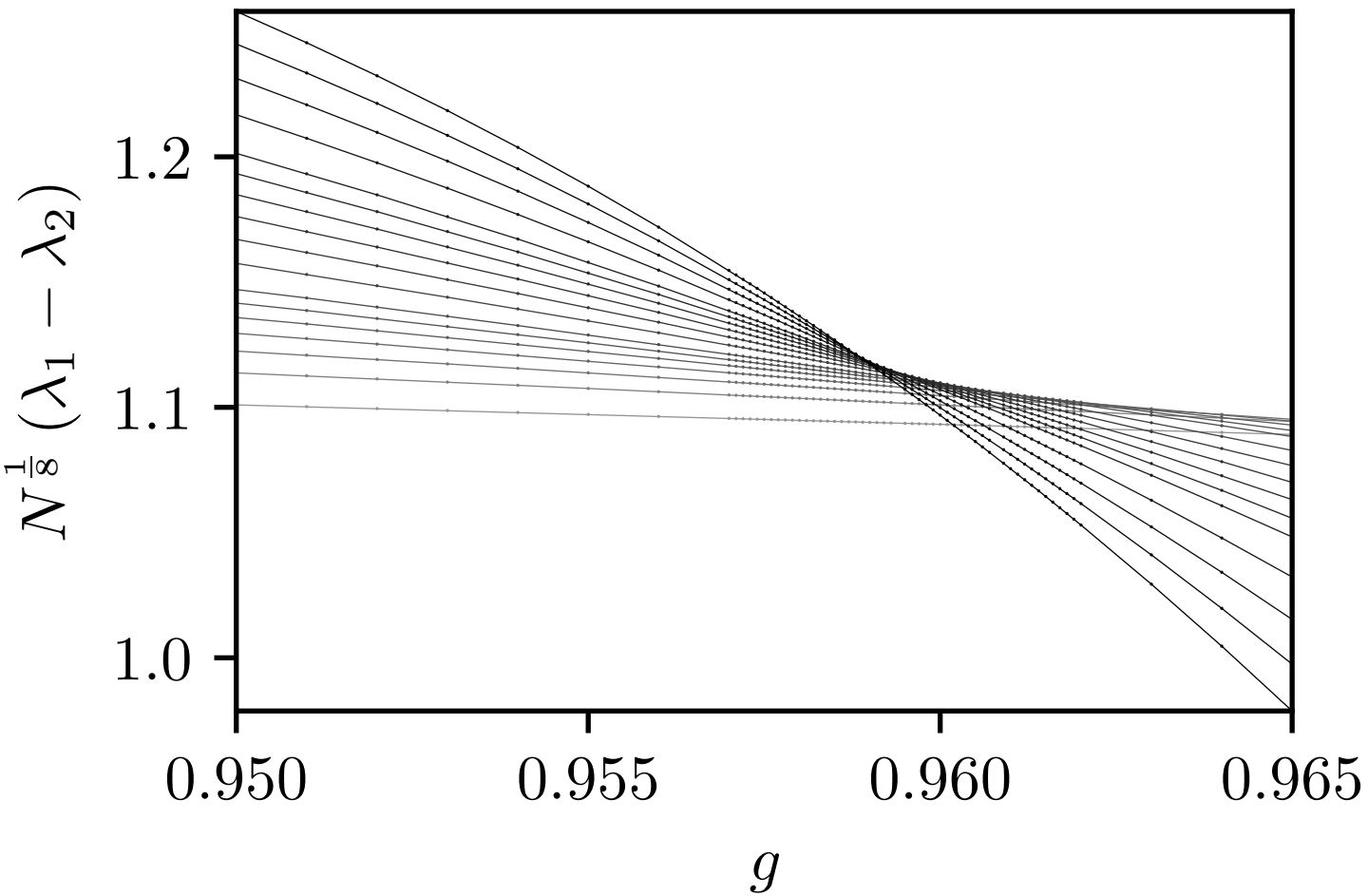
# Transverse field Ising model

- Universal quantities only depend on symmetry and dimensionality
- $\hat{H}$ has $\mathbb{Z}_2$ symmetry (reflection along $z$), same as TFIM
- Critical point of TFIM is known to be in 2D Ising universality class:[1]
  - $\nu = 1$, $\beta = 1/8$, $\gamma = 7/4$

[1] M. Newman and G. Barkema, **Monte Carlo methods in statistical physics**, (Oxford University Press: New York, USA, 1999).

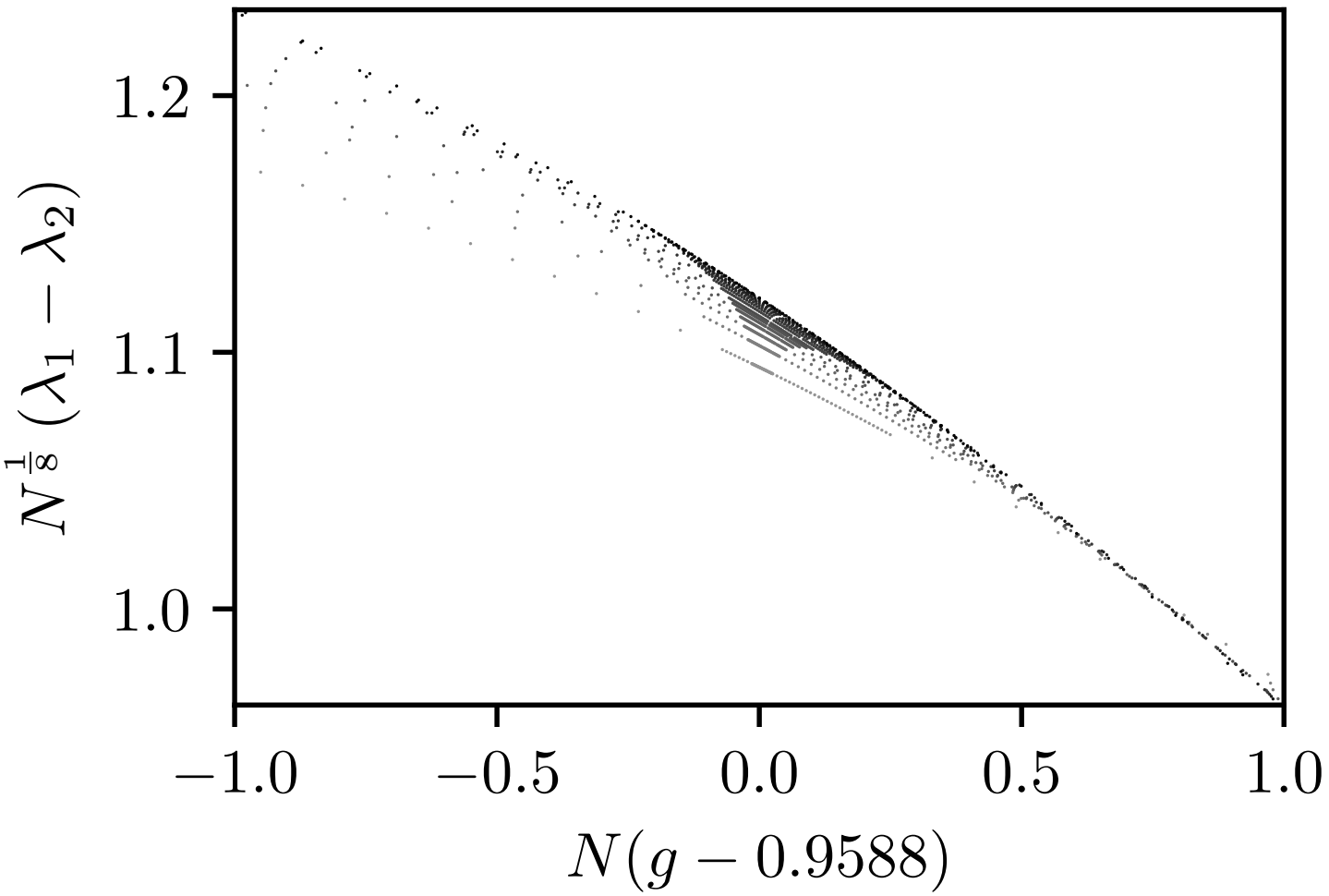
# Schmidt gap

- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



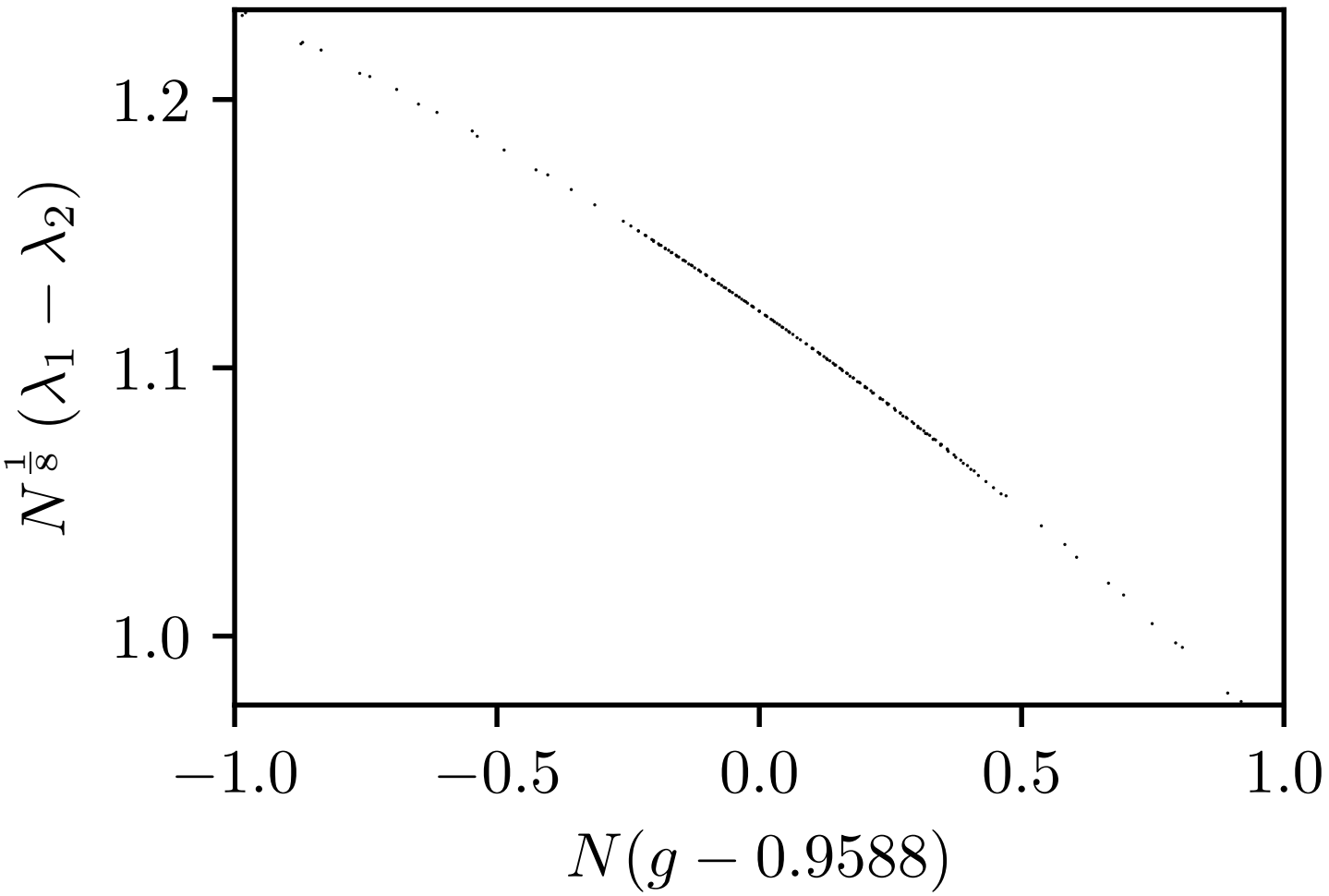
# Schmidt gap

- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



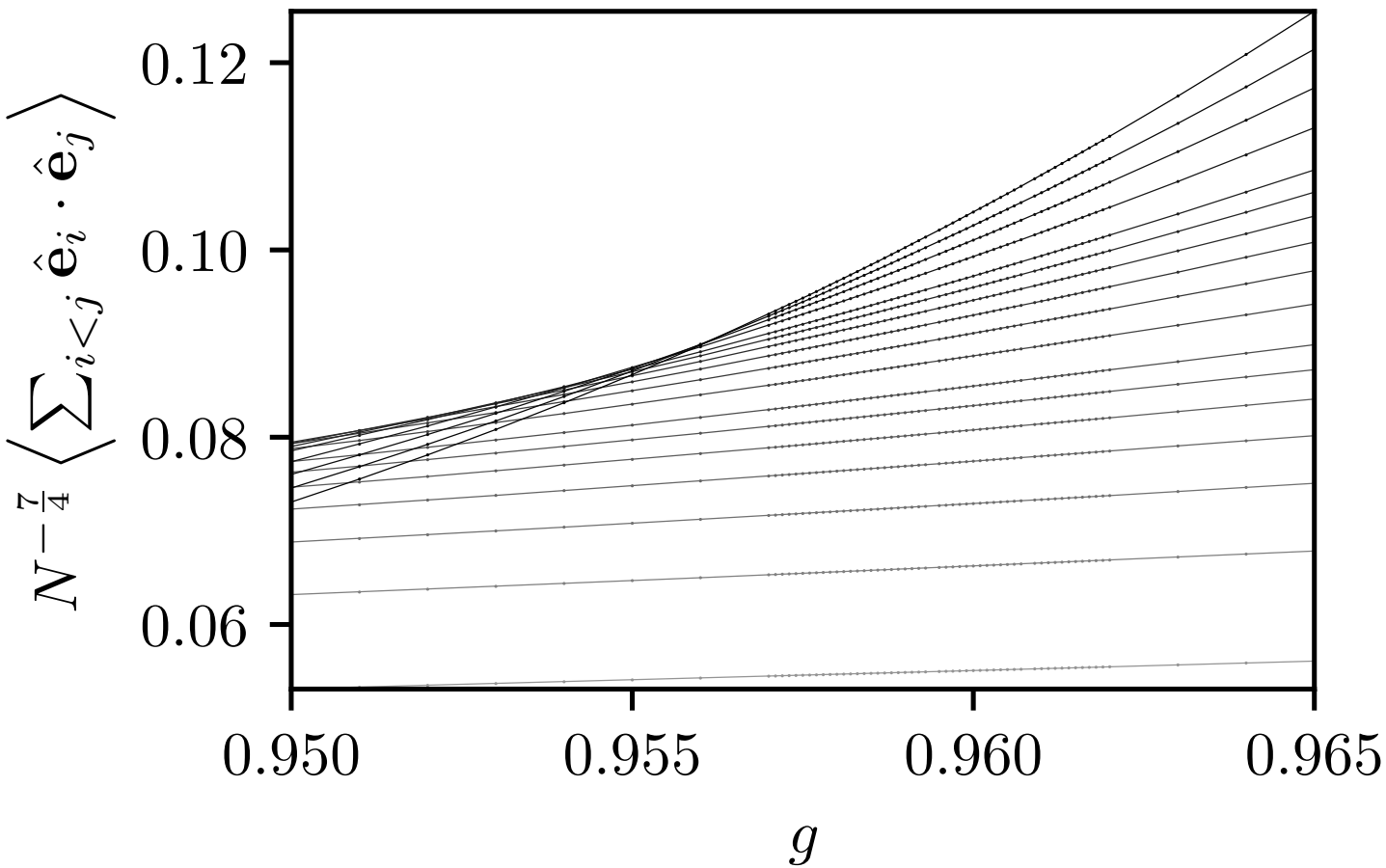
# Schmidt gap

- All interactions, $\ell_{\max} = 2$, $N = 112$ to $144$



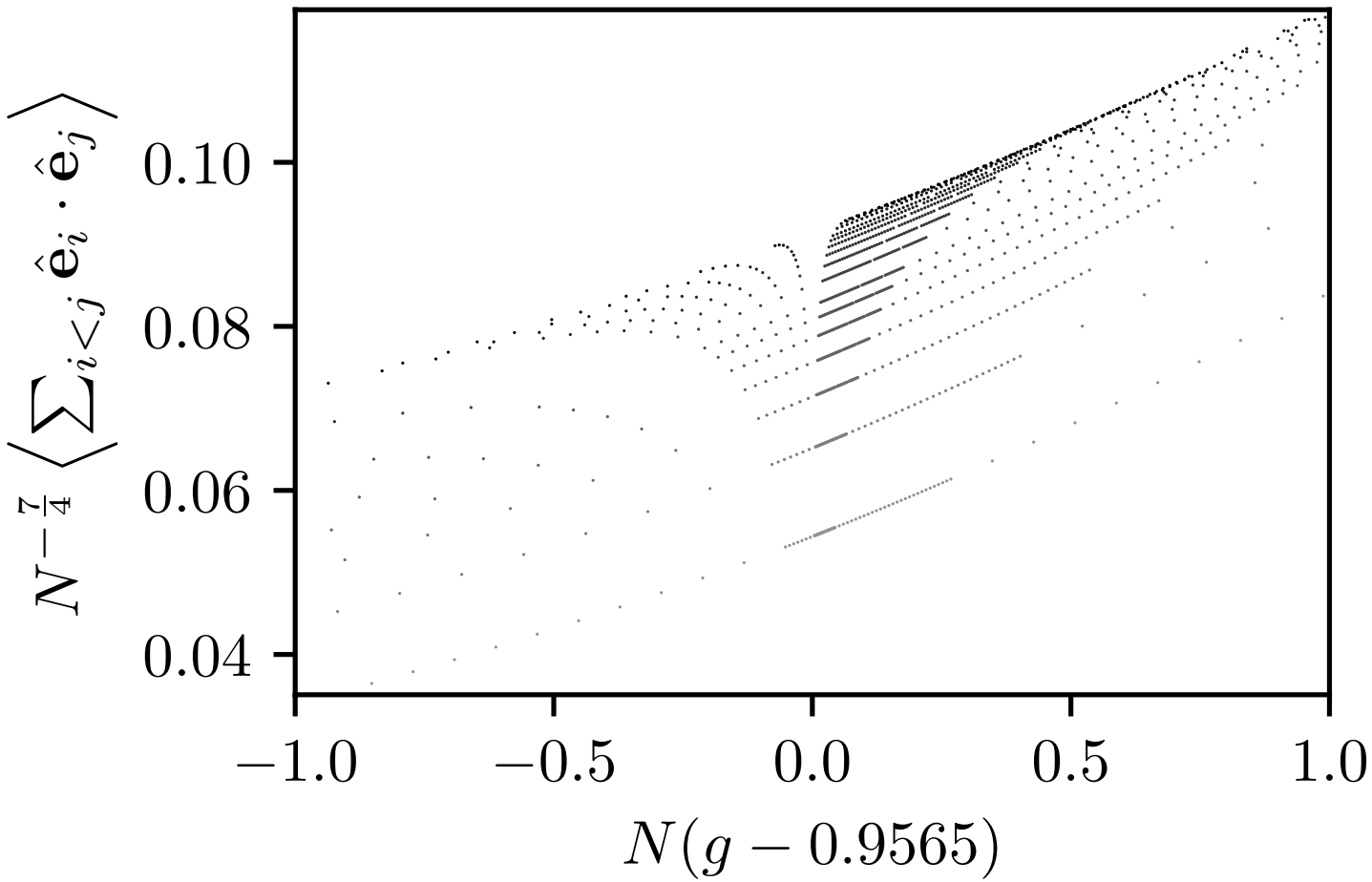
# Orientational correlation

- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



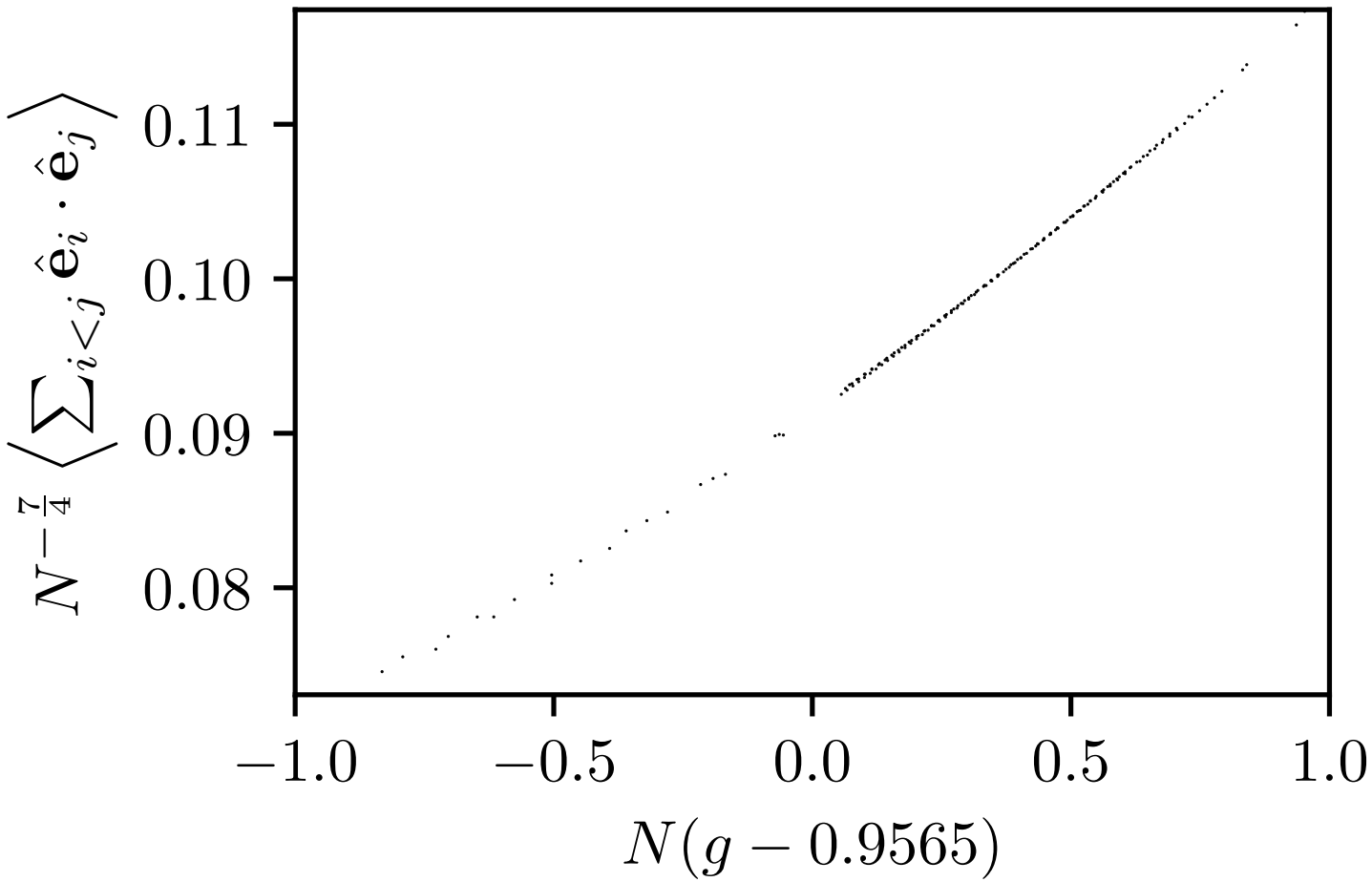
# Orientational correlation

- All interactions, $\ell_{\max} = 2$, $N = 8$ to $144$



# Orientational correlation

- All interactions, $\ell_{\max} = 2$, $N = 112$ to $144$



# Critical parameter

- The value of the critical parameter $g_{\mathrm{c}}$ is non-universal
- In the case of all interactions and $\ell_{\max} = 2$, we have two estimates:
  - $0.9588$ from Schmidt gap
  - $0.9565$ from orientational correlation
- Likely not in the scaling limit at 144 rotors

# Summary

- Simple model of confined dipolar rotors exhibits order–disorder phase transition
- Same universality class (2D Ising) as phase transition in TFIM

- Location of critical point is roughly known
- Larger finite size calculations necessary for complete data collapse

# Acknowledgements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