

# Challenging energy-speed limits in antiferromagnets

Johan H. Mentink

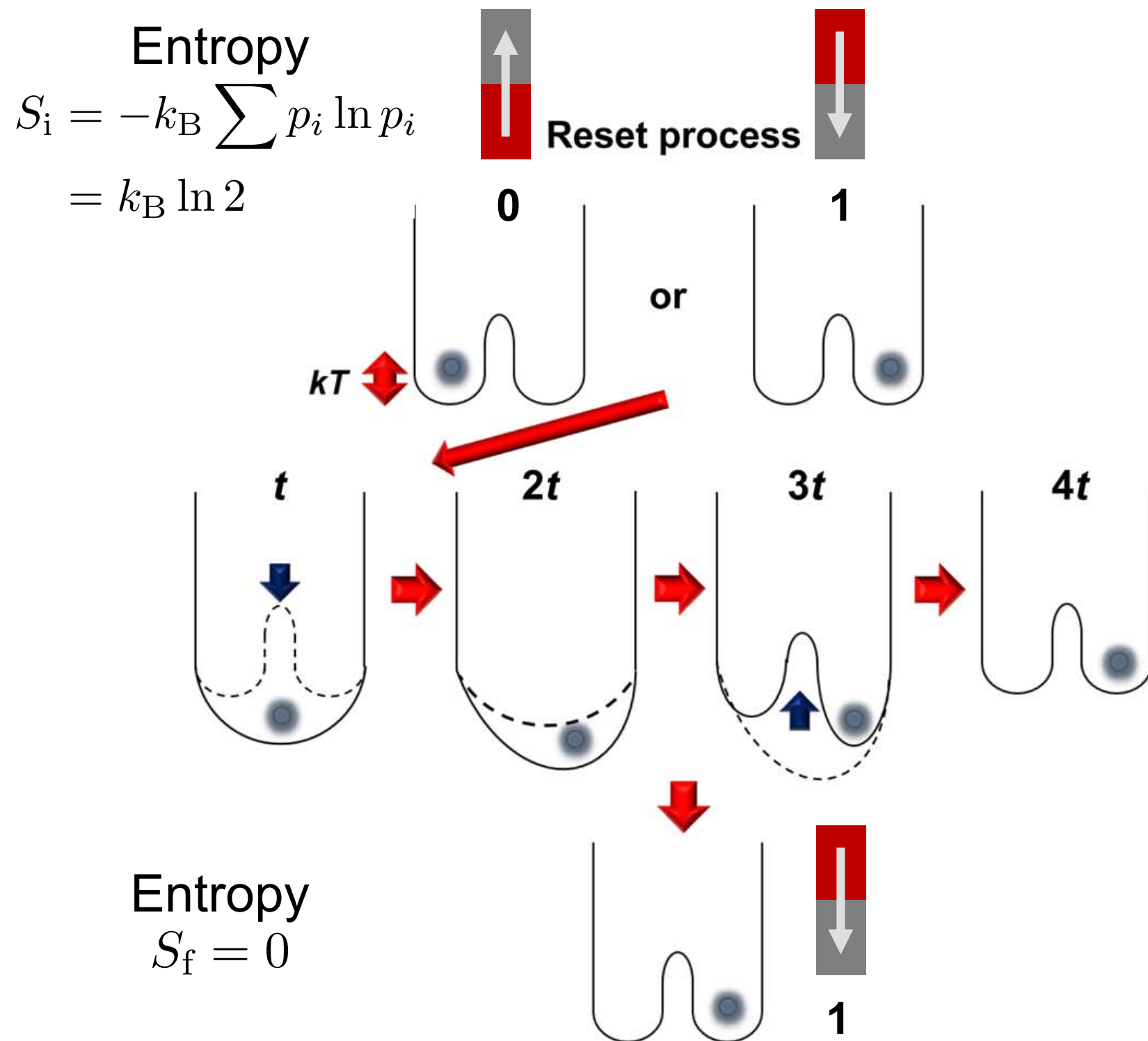
**Institute for Molecules and Materials**

**Radboud University**



SPICE workshop “Ultrafast Antiferromagnetic Writing”, May 09, 2022

# Thermodynamics of computation



$$\Delta E_{\text{int}} = Q + W = 0$$

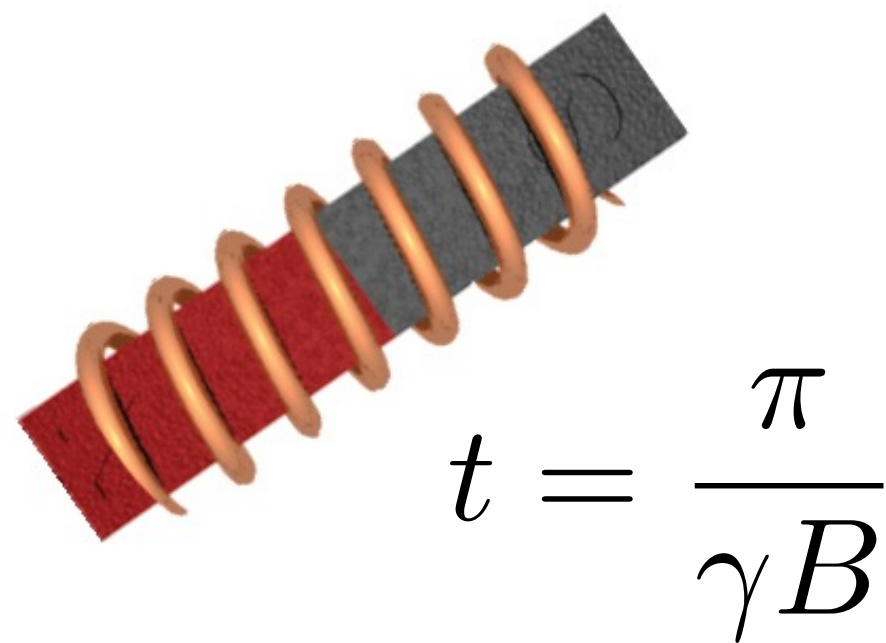
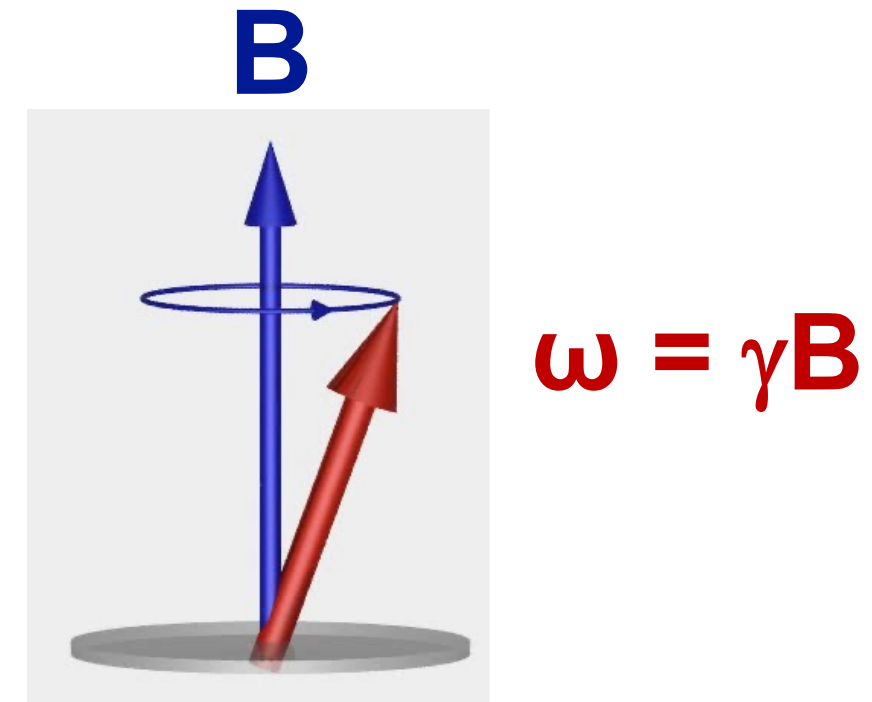
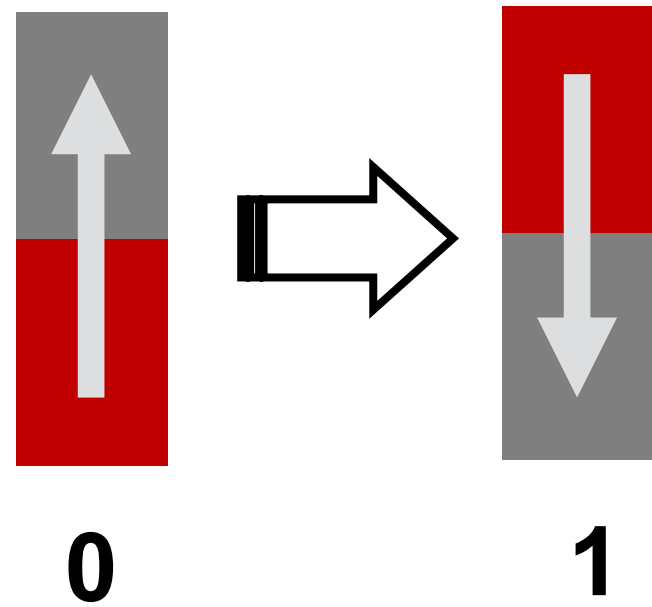
$$W_{\text{min}} = -Q = -T\Delta S$$

## Landauer limit

$$W_{\text{min}} = k_B T \ln 2$$

$$\sim \text{zJ} = 10^{-21} \text{ Joule}$$

# Energy–time dilemma



## Quantum speed limit

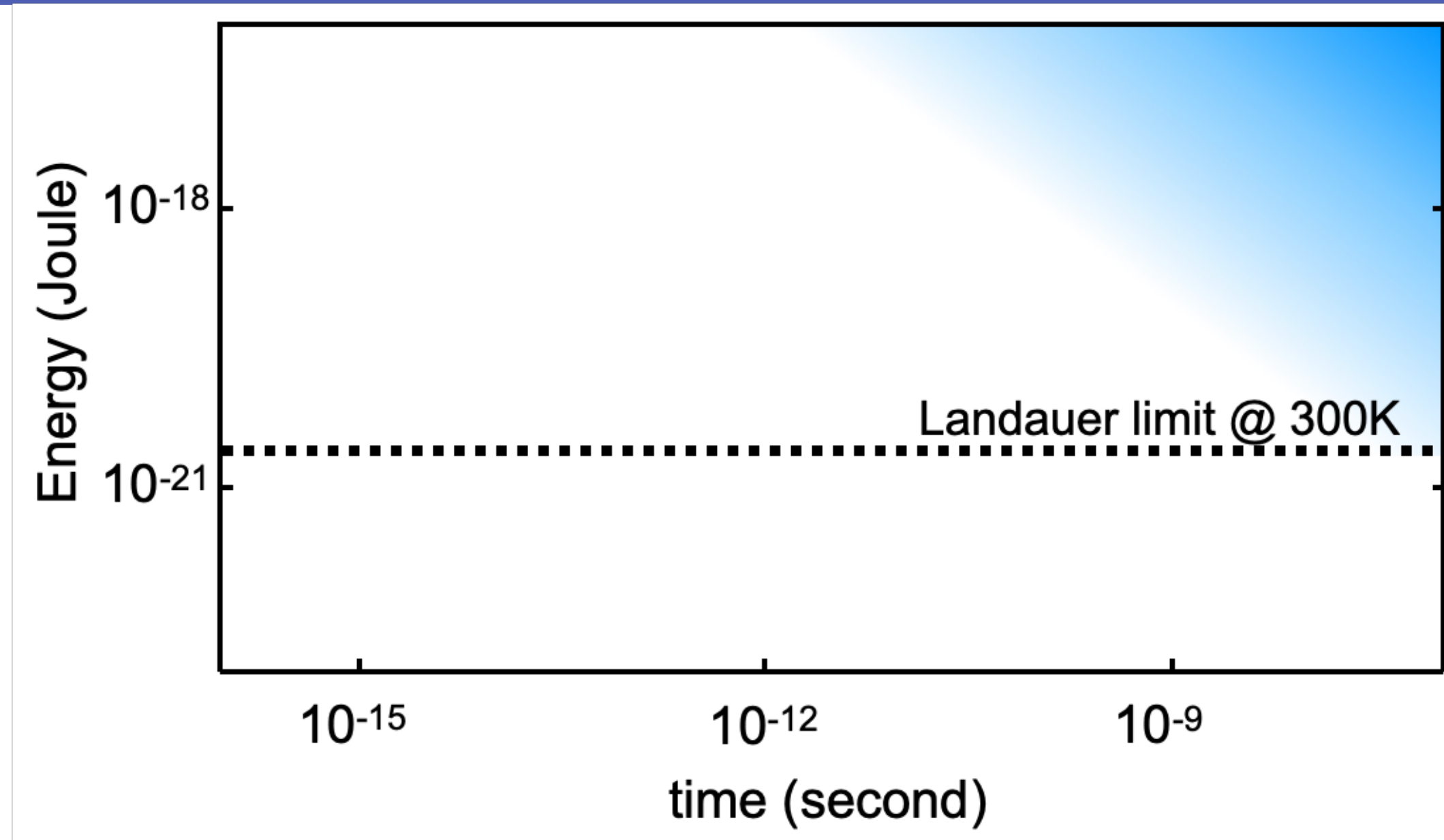
$$t \geq h/4E$$

$$\gamma = 2 \frac{\mu_B}{\hbar} \quad E = \mu_B B$$

S. Rijmer, B.Sc. thesis (2019)

Margolus and Levitin, Phys D. 120, 188 (1998)

# Physical laws

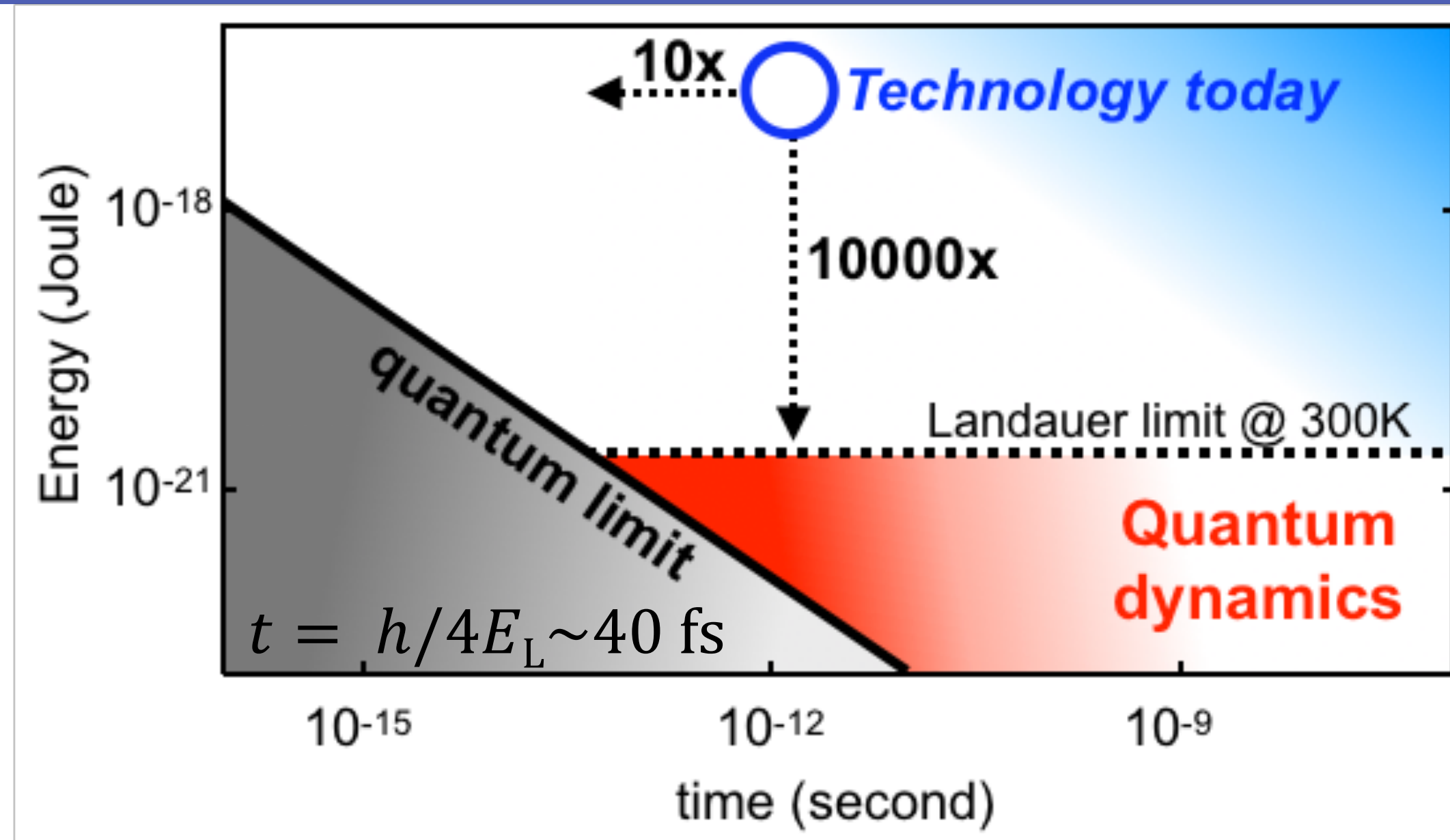


*Quantum speed limit*

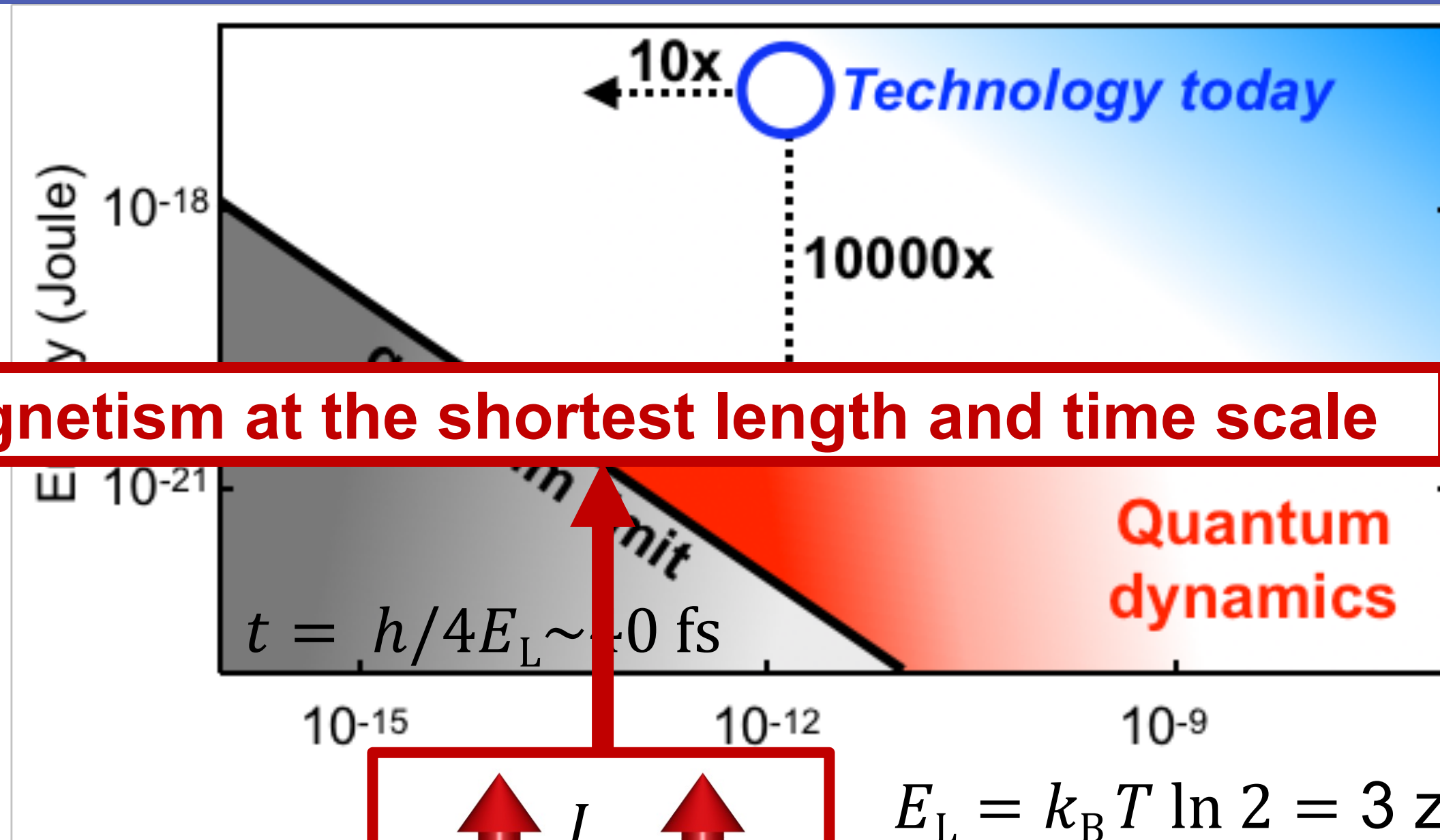
Margolus and Levitin, Phys D. 120, 188 (1998)

Landauer, IBM J Res Dev (1961)

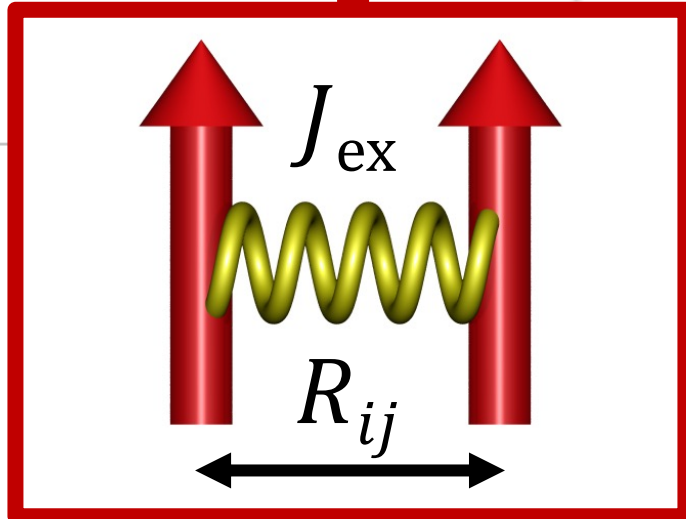
# Physical laws



# Physical laws



**Magnetism at the shortest length and time scale**



$$E_L = k_B T \ln 2 = 3 \text{ zJ} = 25 \text{ meV}$$

$$E_{\text{ex}} = J_{\text{ex}} \vec{S}_1 \vec{S}_2 \sim 25 \text{ meV}$$

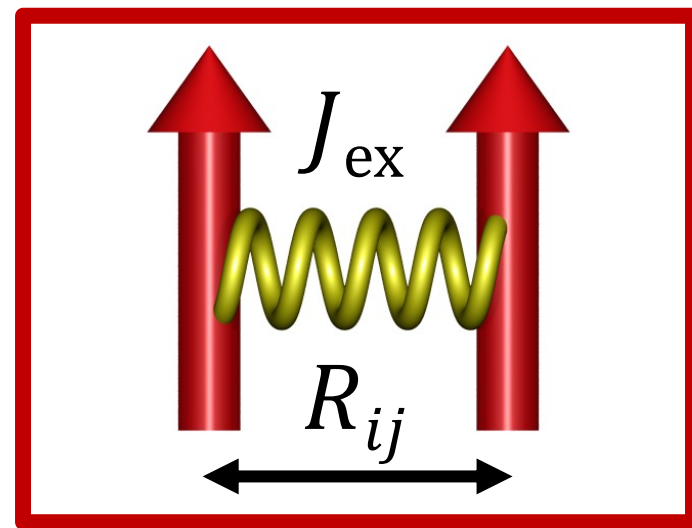
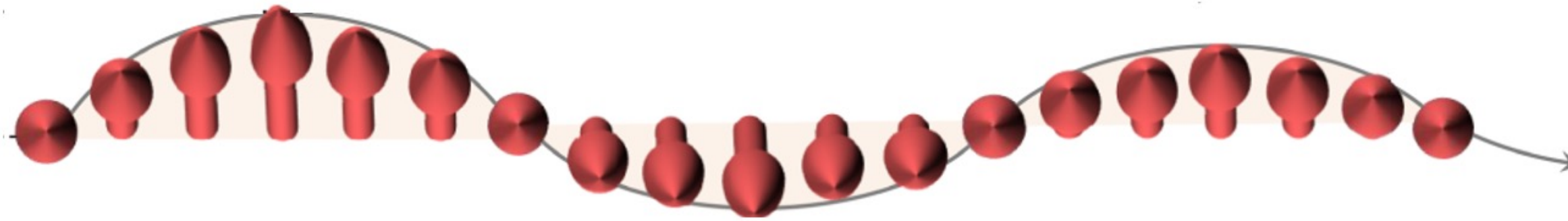
Interaction range  $R_{ij} \sim \text{nm}$

Quantum speed limit

Margolus and Levitin, Phys D. 120, 188 (1998)

Landauer, IBM J Res Dev (1961)

# The smallest and fastest magnetic waves



Wavelength  $\lambda \rightarrow R_{ij} \sim 5\text{\AA}$

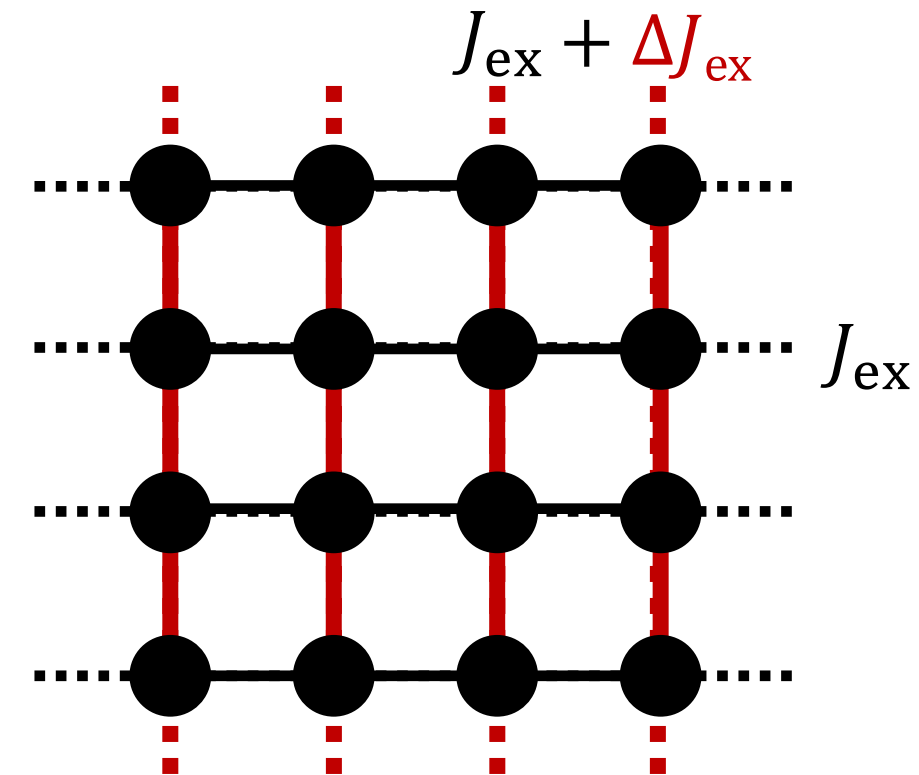
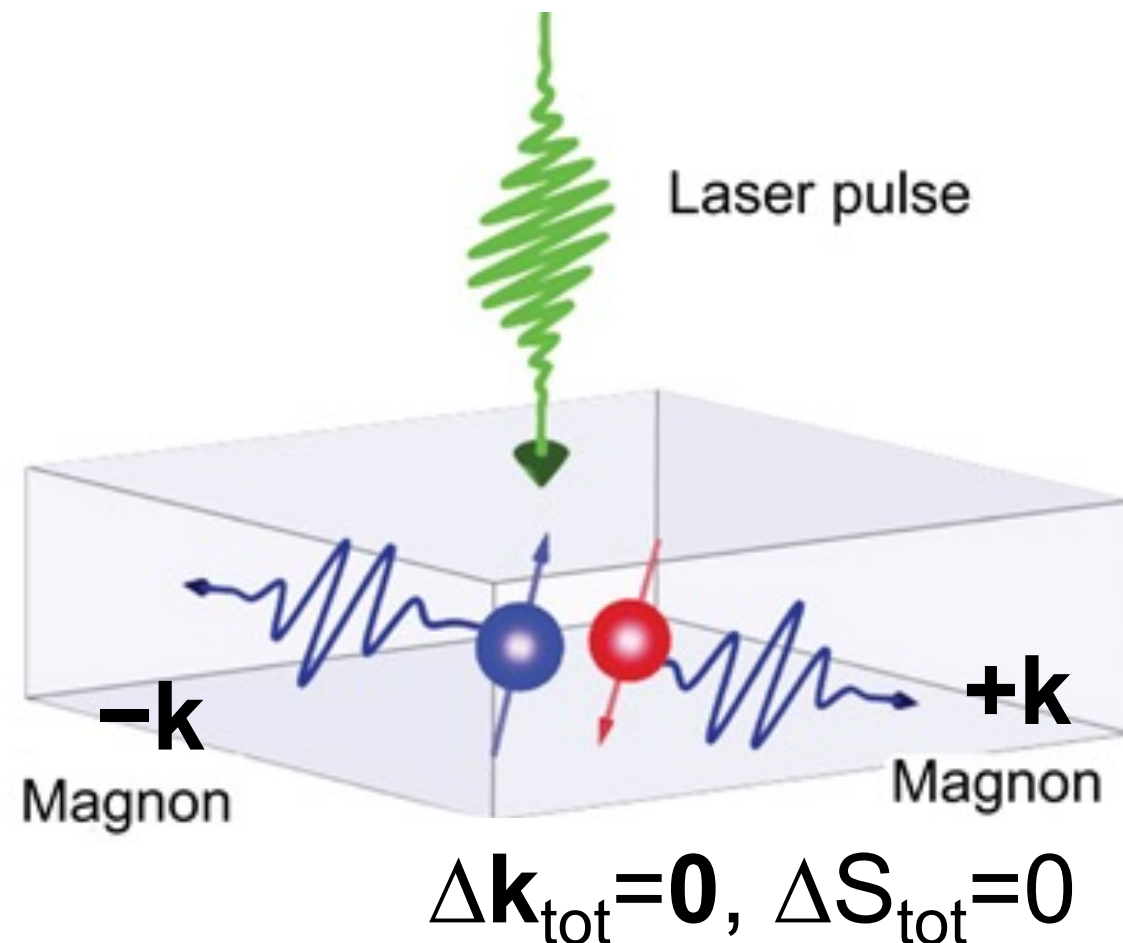
$\hbar\omega \rightarrow E_{\text{ex}} = J_{\text{ex}} \vec{S}_1 \vec{S}_2 \sim 25 \text{ meV}$

Coherent: almost no dissipation

# Excitation of the smallest and fastest magnons

$$\hat{H} = J_{\text{ex}} \sum_{i,\delta} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\delta}$$

$$\Delta\hat{H}(t) = \Delta J_{\text{ex}} f(t) \sum_{i,\delta} (\hat{e} \cdot \vec{\delta})^2 \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\delta}$$



$$\hat{H} = \sum_{\mathbf{k}} \omega_{\mathbf{k}} (\hat{\alpha}_{\mathbf{k}}^{\dagger} \hat{\alpha}_{\mathbf{k}} + \hat{\alpha}_{-\mathbf{k}}^{\dagger} \hat{\alpha}_{-\mathbf{k}} + 1)$$

$$\Delta\hat{H}(t) = f(t) \sum_{\mathbf{k}} V_{\mathbf{k}} (\hat{\alpha}_{\mathbf{k}}^{\dagger} \hat{\alpha}_{-\mathbf{k}}^{\dagger} + \hat{\alpha}_{\mathbf{k}} \hat{\alpha}_{-\mathbf{k}})$$

$V_{\mathbf{k}}$  dominant at  $\mathbf{k} = (0, \pm\pi)$

J. Zhao et al., PRL 93, 107203 (2004)

Bossini et al., Nat. Commun. 7, 10645 (2016)

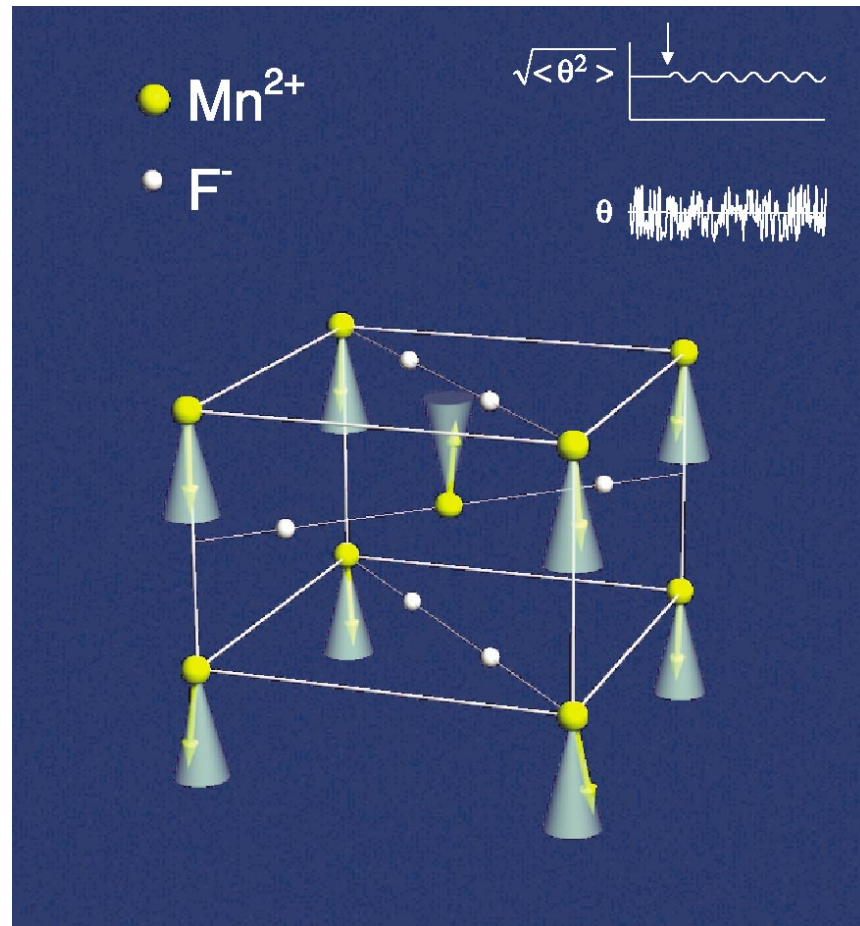
D. Bossini, .. ,O. Gomonay, ..., J.H. Mentink, .. et al., PRB 100, 024428 (2019)

Spontaneous RS: Fleury and Loudon, Phys Rev. 1968

Ultrafast: Mentink et al., Nat. Commun. 2015

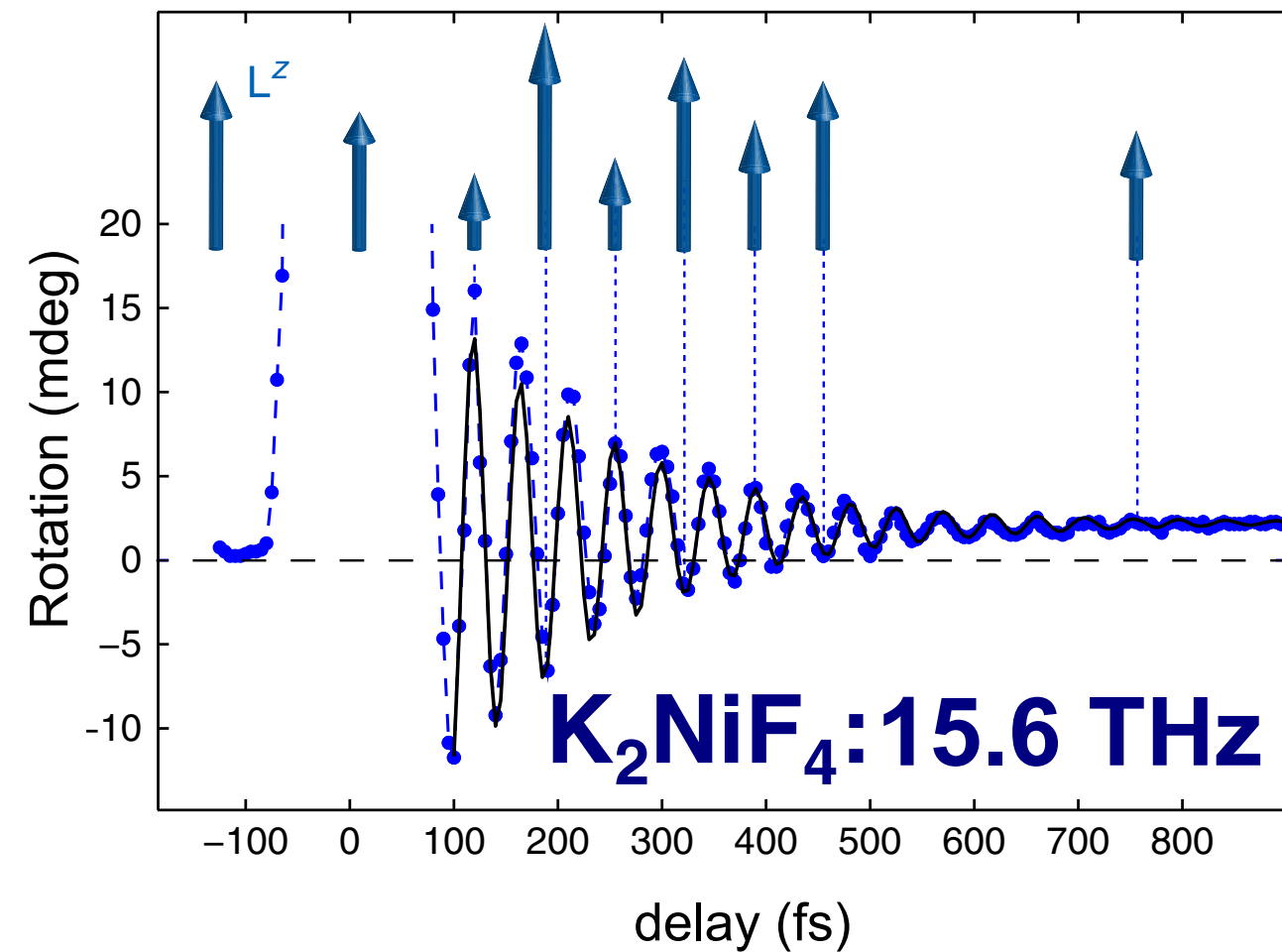


# Time-resolved dynamics of the smallest and fastest magnons



Magnon squeezing

Zhao, Merlin et al.,  
*PRL* 2004, *PRB* 2006



Coherent *longitudinal* dynamics  
Magnon entanglement

D. Bossini et al.,  
*Nat. Commun.* (2016), *PRB* (2019)

# Outline

## 1. Magnon-pair physics

J. Zhao et al., PRL **93**, 107203 (2004);

D. Bossini et al, Nat. Commun. **7**, 10645 (2016);

D. Bossini, .. , O. Gomonay, .., J.H. Mentink et al., PRB **100**, 024428 (2019)

## 2. Supermagnonic propagation of magnon pairs

G. Fabiani, M.D. Bouman, J.H. Mentink

Phys. Rev. Lett. **127**, 097202 (2021)

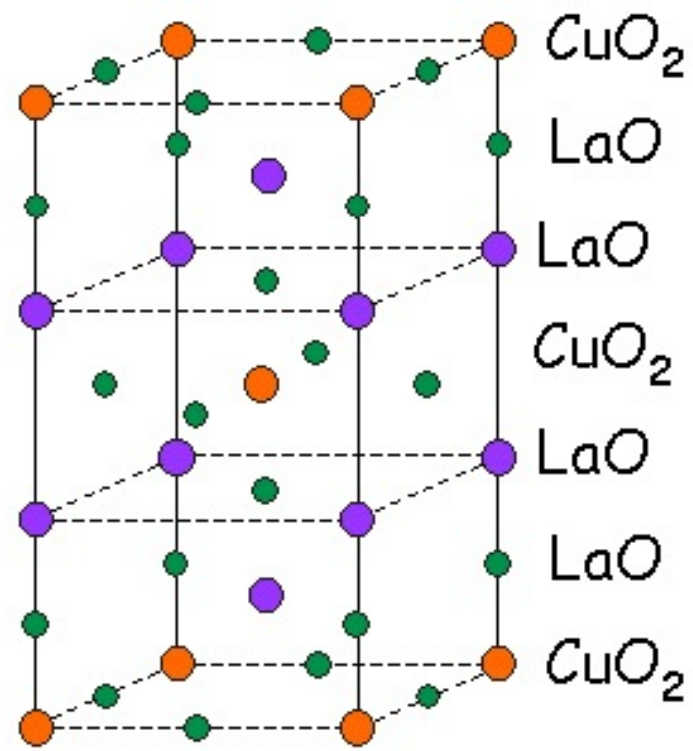
## 3. Semi-classical approach to nonlinear magnon-pair dynamics

G. Fabiani and J.H. Mentink

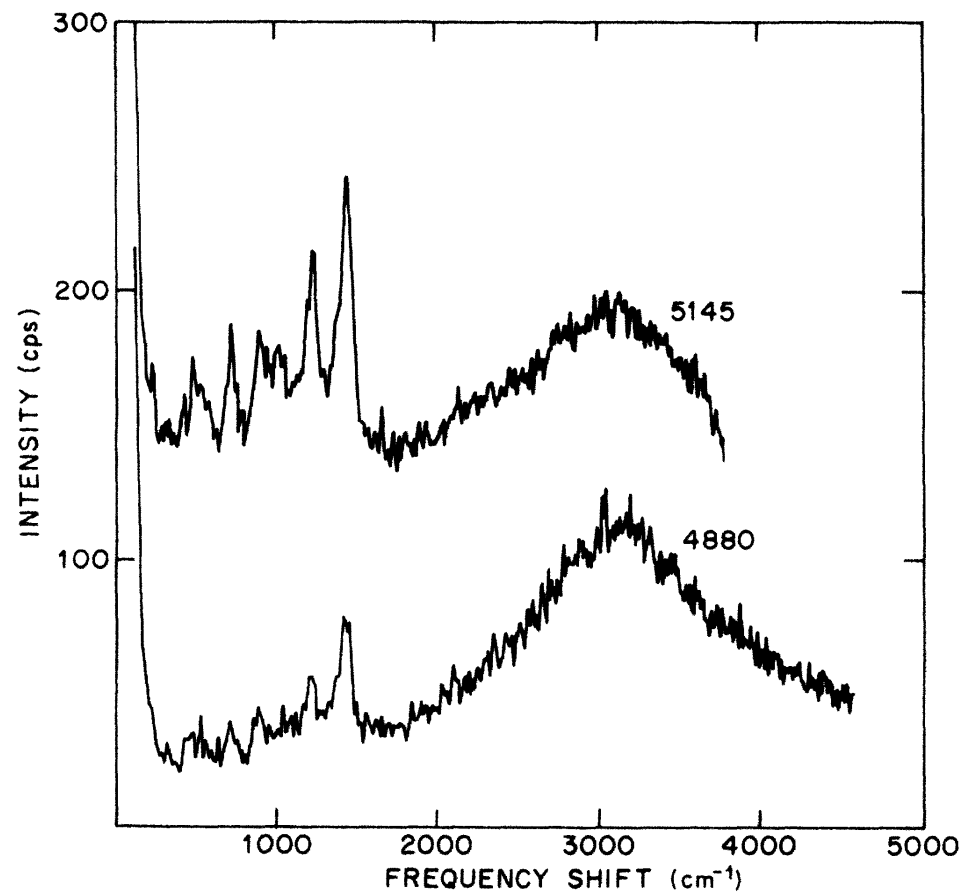
Appl. Phys. Lett. **120**, 152402 (2022)

# Challenges at the Edge of the Brillouin Zone

parent compounds  
cuprates  
(high-T<sub>c</sub>, Nobelprize 1987)  
La<sub>2</sub>CuO<sub>4</sub>



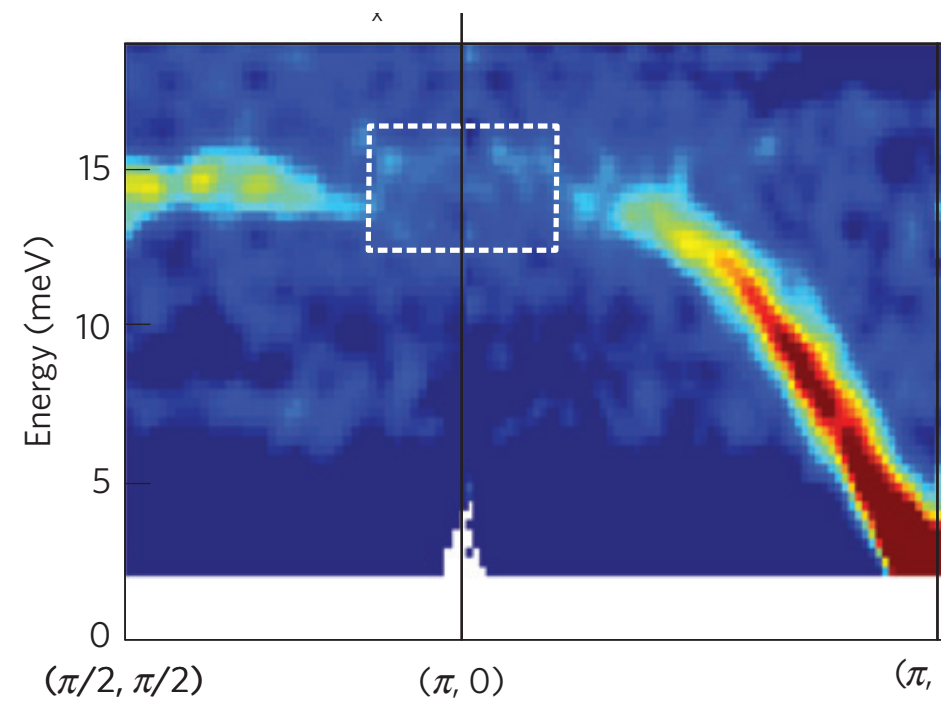
metal-organic compound  
Cu(DCOO)<sub>2</sub> · 4D<sub>2</sub>O (CFTD)



S=1/2 Heisenberg antiferromagnet in 2D

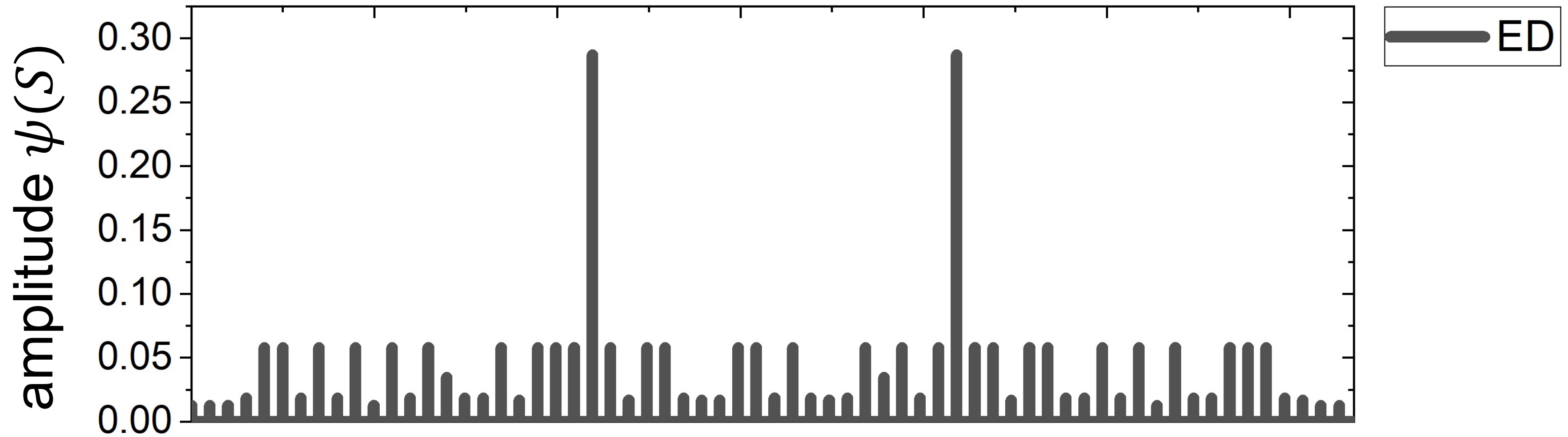
$$\hat{H} = J_{\text{ex}} \sum_{i,\delta} \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\delta}$$

Lyons et al., PRB 37, 2353 (1988)



B. Dalla Piazza et al., Nature Physics 11, 62 (2015)  
H. Shao, et al., PRX 7 041072 (2017)

# The many-body wavefunction

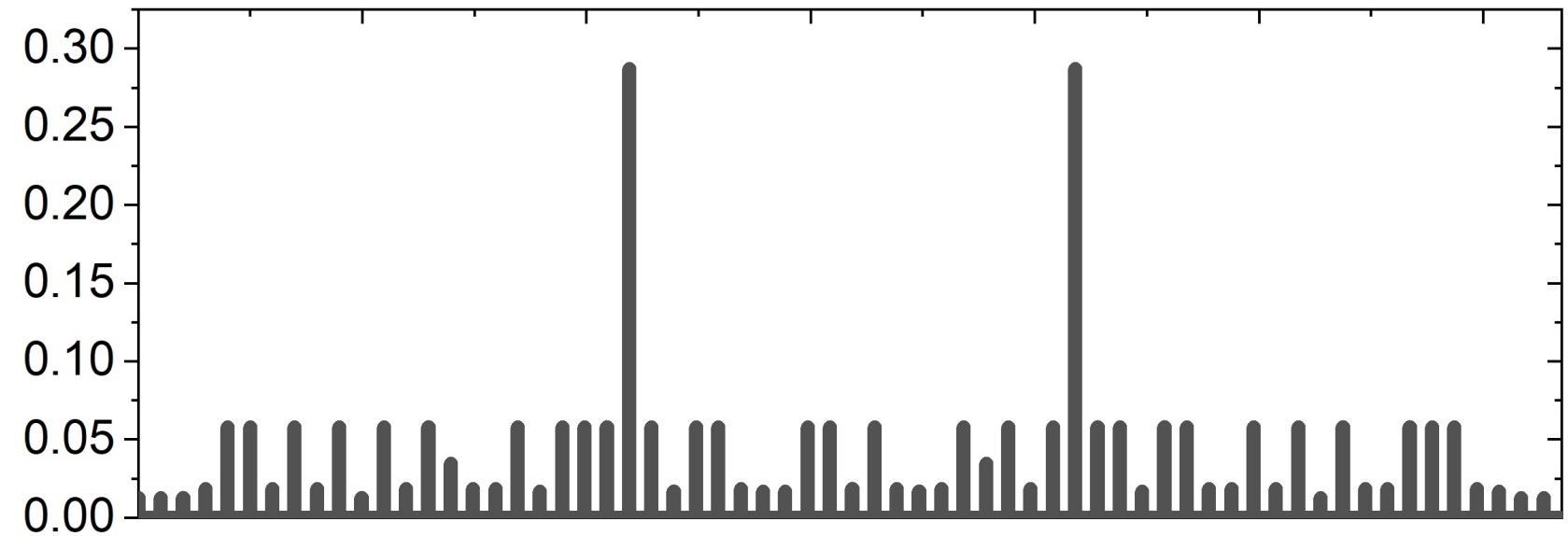
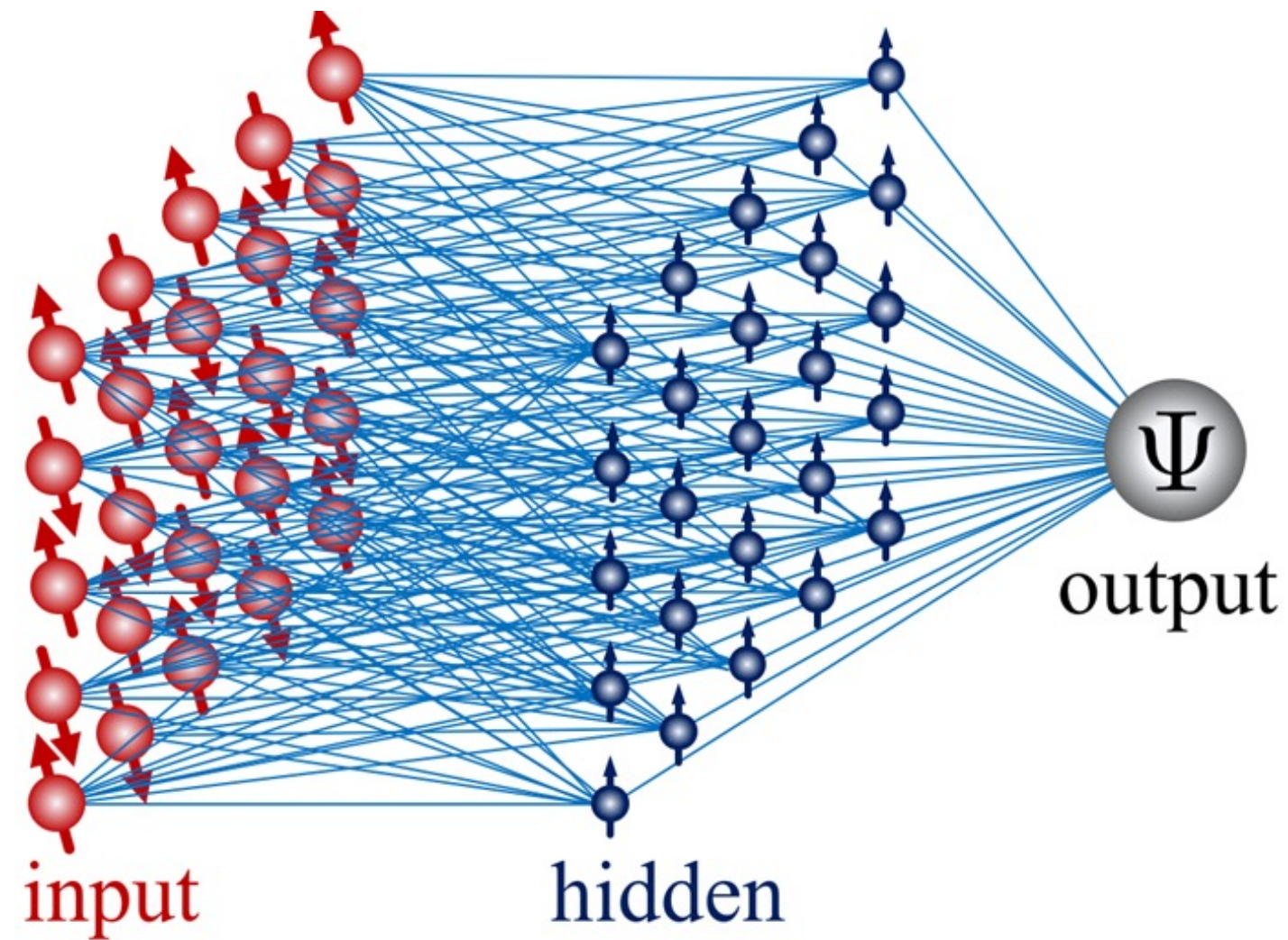


All possible quantum states  $|S_1\rangle \dots |S_{2^N}\rangle$

$2^N$  states

$$|S\rangle = |\underbrace{\uparrow\downarrow\uparrow \dots \uparrow}_{N \text{ spins}}\rangle$$

# Wave function as artificial neural network



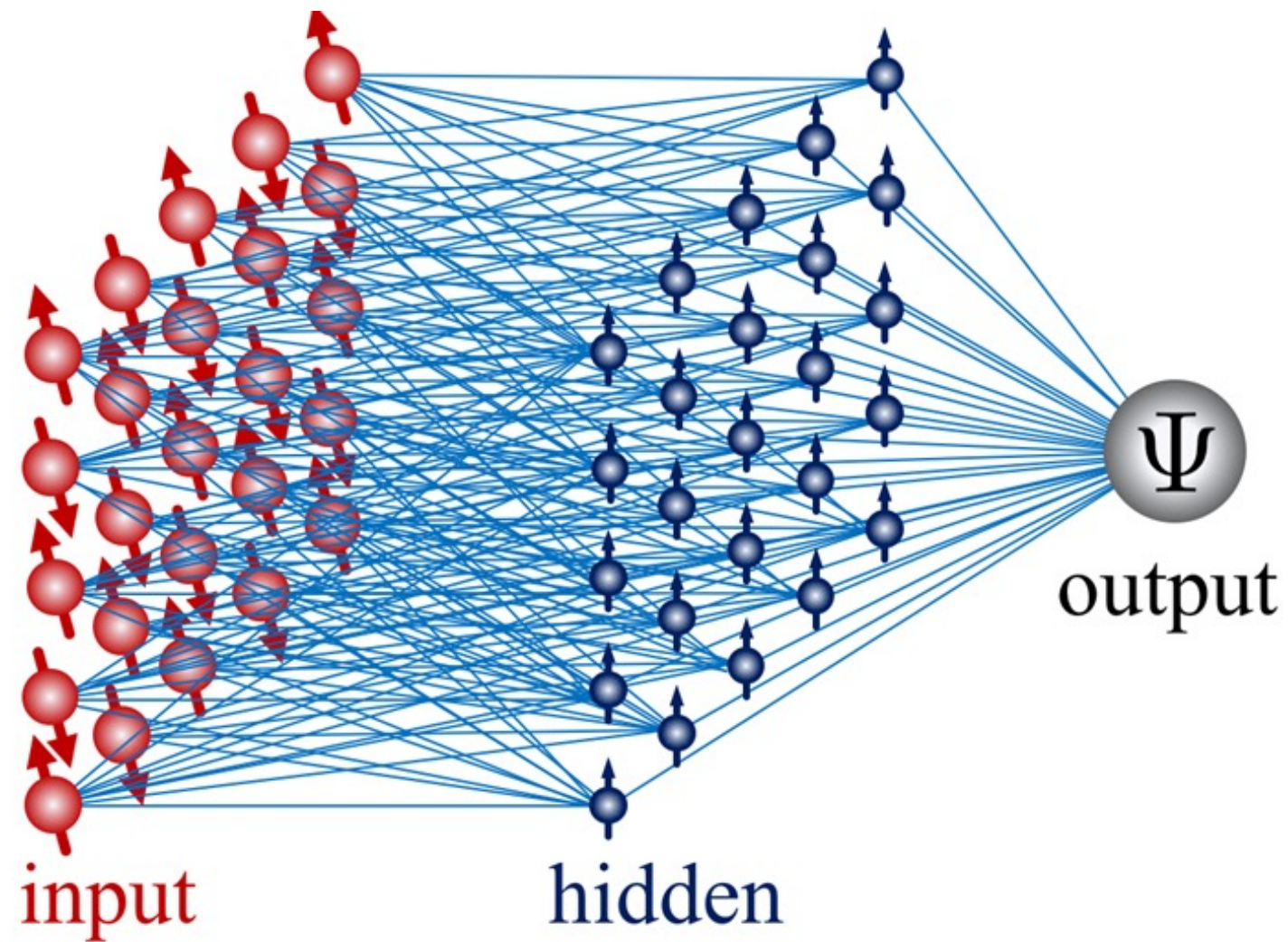
All possible quantum states  $|S_1\rangle \dots |S_{2N}\rangle$

$$|S\rangle = |\uparrow\downarrow\uparrow \dots \uparrow\rangle$$

- Universal function approximation theorem
- Reduction from  $2^N$  to  $\alpha N$  parameters
- Much reduced limits on simulation time / system size

G. Carleo, M. Troyer  
Science 355, 602 (2017)

# Wave function as artificial neural network



$$|S\rangle = |\uparrow\downarrow\uparrow \cdots \uparrow\rangle$$

G. Carleo, M. Troyer  
Science 355, 602 (2017)

## Neural-network quantum states (NQS)

$$\psi_{\mathcal{W}}(S) = \sum_{\{h_i\}} e^{\sum_j a_j s_j^Z + \sum_i b_i h_i + \sum_{ij} w_{ij} s_i^Z h_j}$$

Restricted Boltzmann Machine

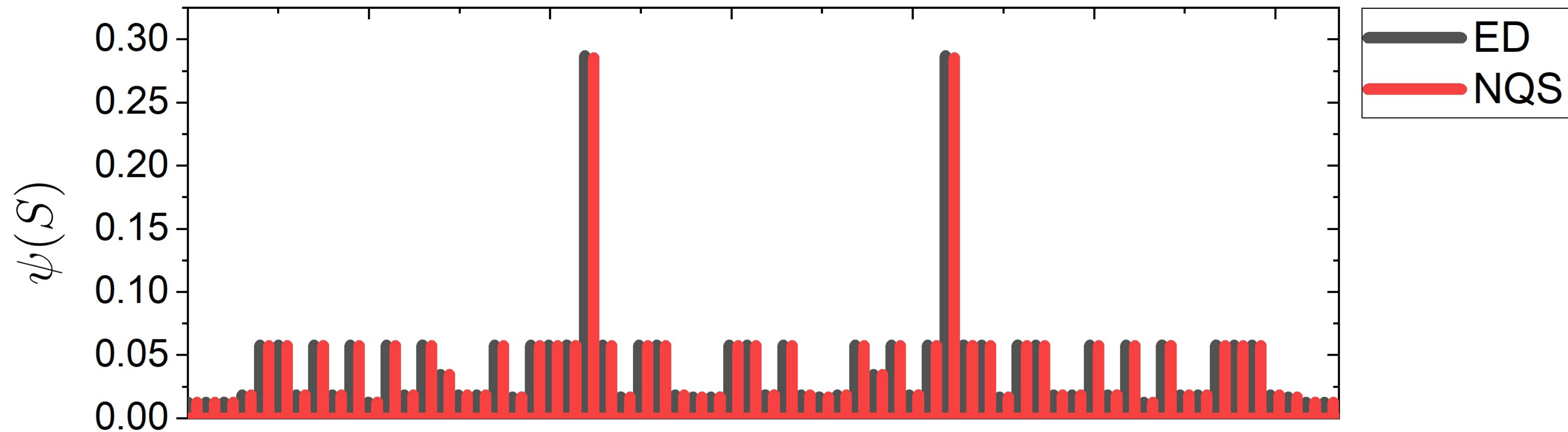
## Optimization

Ground state: minimize  $\|(\hat{\mathcal{H}} - E)\psi_{\mathcal{W}}\|$

Dynamics: minimize  $\|i\partial_t\psi_{\mathcal{W}}(t) - \hat{\mathcal{H}}(t)\psi_{\mathcal{W}}(t)\|$

“unsupervised” learning from samples

# Ground state NQS vs Exact Diagonalization (ED)



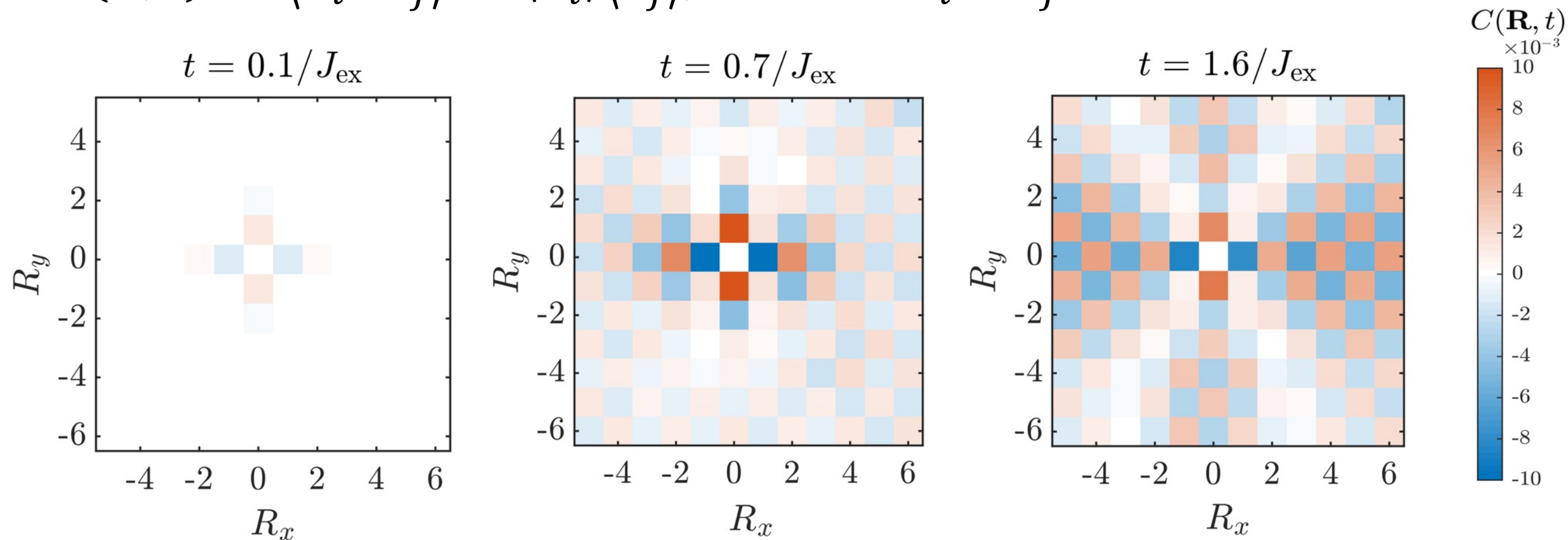
Network with  $M = \alpha N = 64$  parameters already gives accurate results

# Propagation of fastest magnetic waves

$$\Delta J_{\text{ex}} f(t)$$

$$\Delta \hat{H}(t) = \Delta J_{\text{ex}} f(t) \sum_{i,\delta} (\hat{e} \cdot \vec{\delta})^2 \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\delta}$$

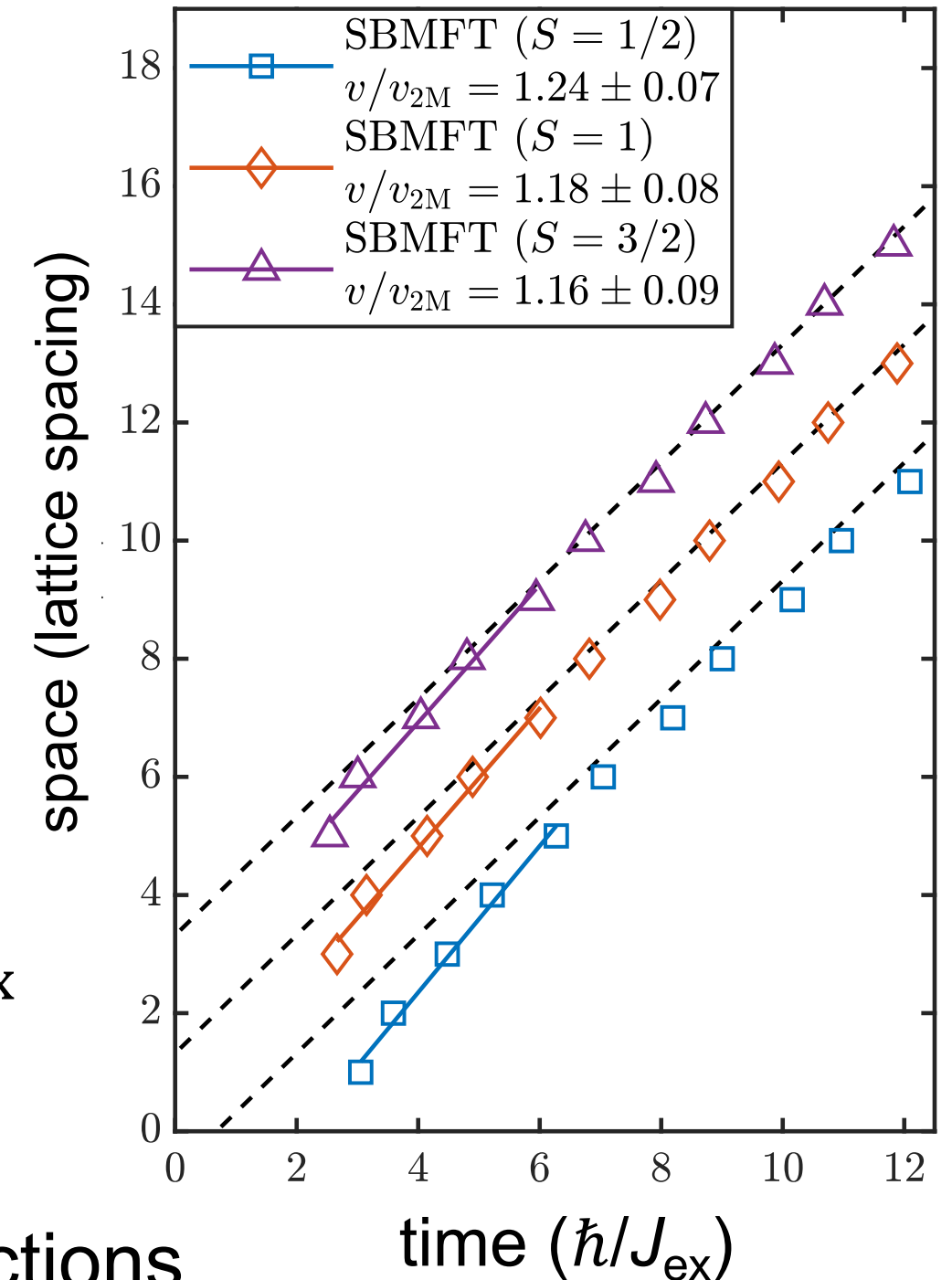
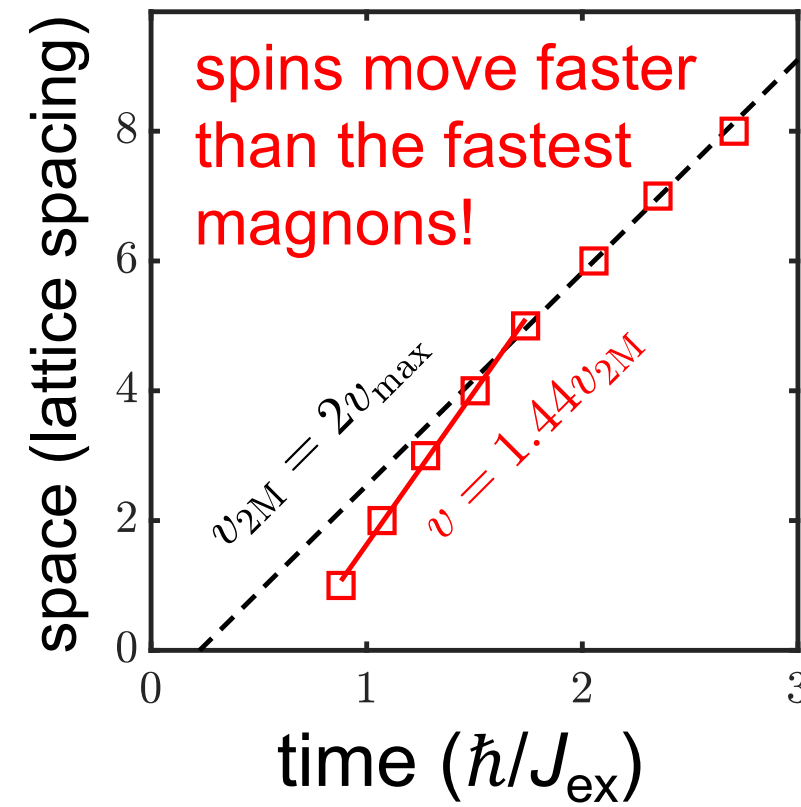
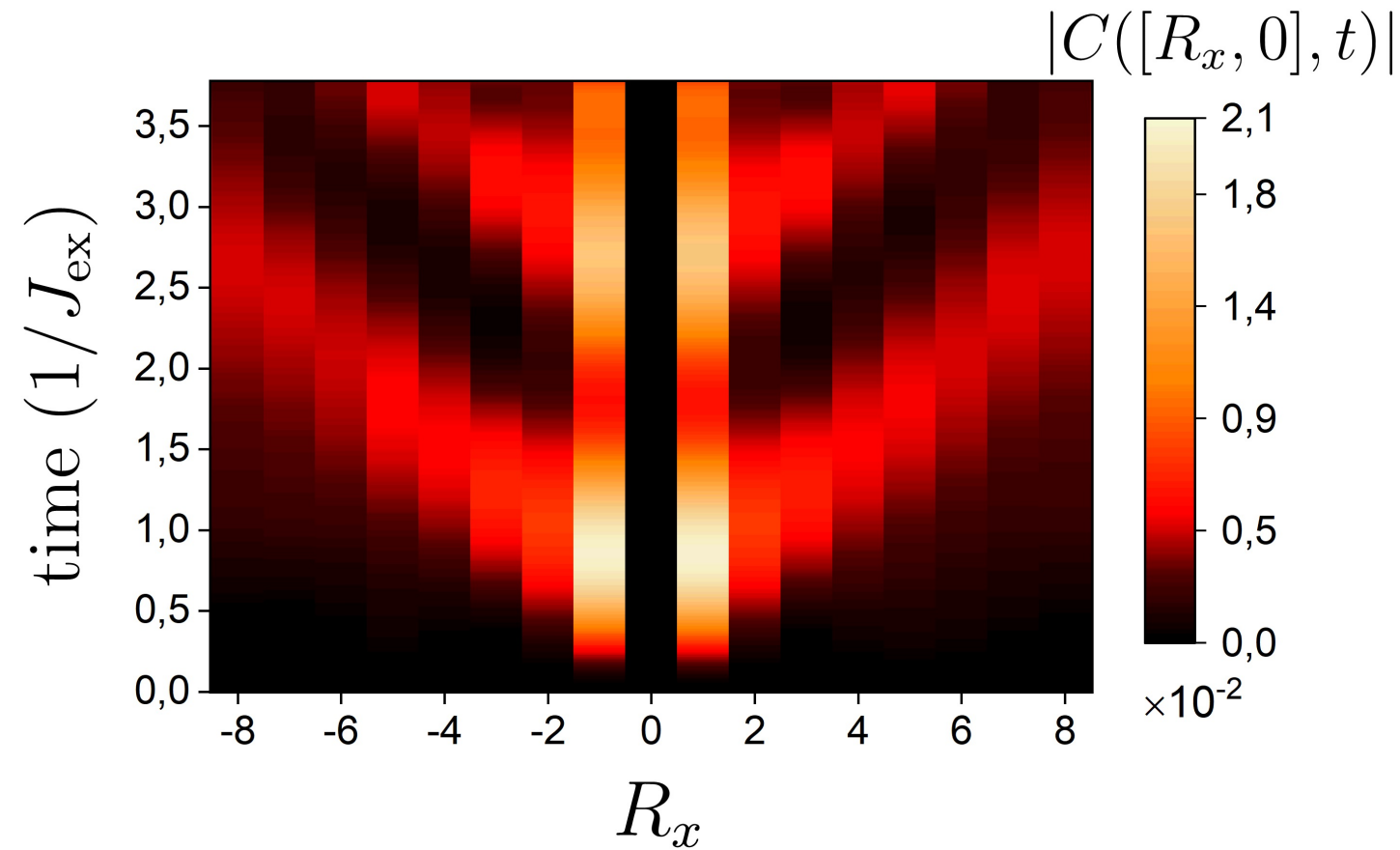
$$C(\mathbf{R}, t) = \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle - \langle \mathbf{S}_i \rangle \langle \mathbf{S}_j \rangle, \quad \mathbf{R} = \mathbf{r}_i - \mathbf{r}_j$$



Anisotropy of propagation determined by symmetry of light-matter interaction



# Supermagnonic propagation



For small  $R_x$  faster

$v(\text{NQS}) \approx 4.71 a J_{\text{ex}}$  **40% higher** than  $v(\text{LSWT}) \approx 3.28 a J_{\text{ex}}$

$\approx 20 \text{ km/s}$  for  $J_{\text{ex}} \approx 6 \text{ meV}$ ,  $a \approx 5 \text{ \AA}$

Consequence of exceptionally strong magnon-magnon interactions

# Outline

## 1. Magnon-pair physics

J. Zhao et al., PRL **93**, 107203 (2004);

D. Bossini et al, Nat. Commun. **7**, 10645 (2016);

D. Bossini, .. , O. Gomonay, .., J.H. Mentink et al., PRB **100**, 024428 (2019)

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Phys. Rev. Lett. **127**, 097202 (2021)

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# Magnon-pair operator algebra

$$\hat{\mathcal{H}} = \sum_k \omega_k \underbrace{(\hat{\alpha}_k^\dagger \hat{\alpha}_k + \hat{\beta}_{-k}^\dagger \hat{\beta}_{-k} + 1)}_{2\hat{K}_k^z} + f(t) \sum_k V_k \left( \underbrace{\hat{\alpha}_k^\dagger \hat{\beta}_{-k}^\dagger}_{\hat{K}_k^+} + \underbrace{\hat{\alpha}_k \hat{\beta}_{-k}}_{\hat{K}_k^-} \right)$$

**SU(2)**

$$[\hat{S}_i^z, \hat{S}_j^\pm] = \pm \hat{S}_i^\pm \delta_{i,j}$$

$$[\hat{S}_i^-, \hat{S}_j^+] = -2\hat{S}_i^z \delta_{i,j}$$

$$\hat{S}_i^2 = (\hat{S}_i^z)^2 + \frac{1}{2} (\hat{S}_i^- \hat{S}_i^+ + \hat{S}_i^+ \hat{S}_i^-) = \text{const}$$

$$(\hat{S}_i^z)^2 + (\hat{S}_i^x)^2 + (\hat{S}_i^y)^2$$

**SU(1,1)**

$$[\hat{K}_k^z, \hat{K}_{k'}^\pm] = \pm \hat{K}_k^\pm \delta_{k,k'}$$

$$[\hat{K}_k^-, \hat{K}_{k'}^+] = 2\hat{K}_k^z \delta_{k,k'}$$

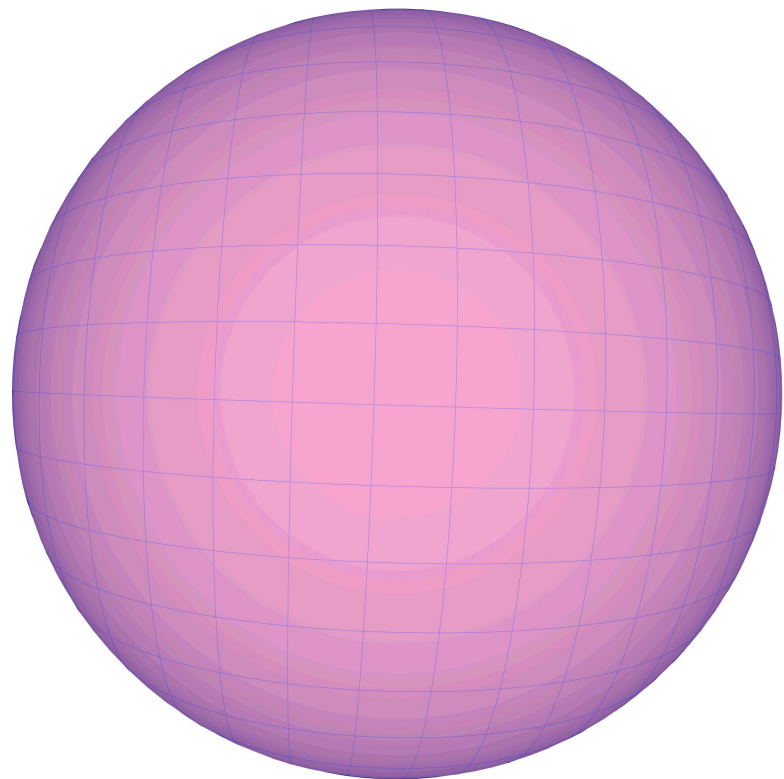
$$\hat{K}_k^2 = -(\hat{K}_k^z)^2 + \frac{1}{2} (\hat{K}_k^- \hat{K}_k^+ + \hat{K}_k^+ \hat{K}_k^-) = \text{const}$$

$$-(\hat{K}_k^z)^2 + (\hat{K}_k^x)^2 + (\hat{K}_k^y)^2$$

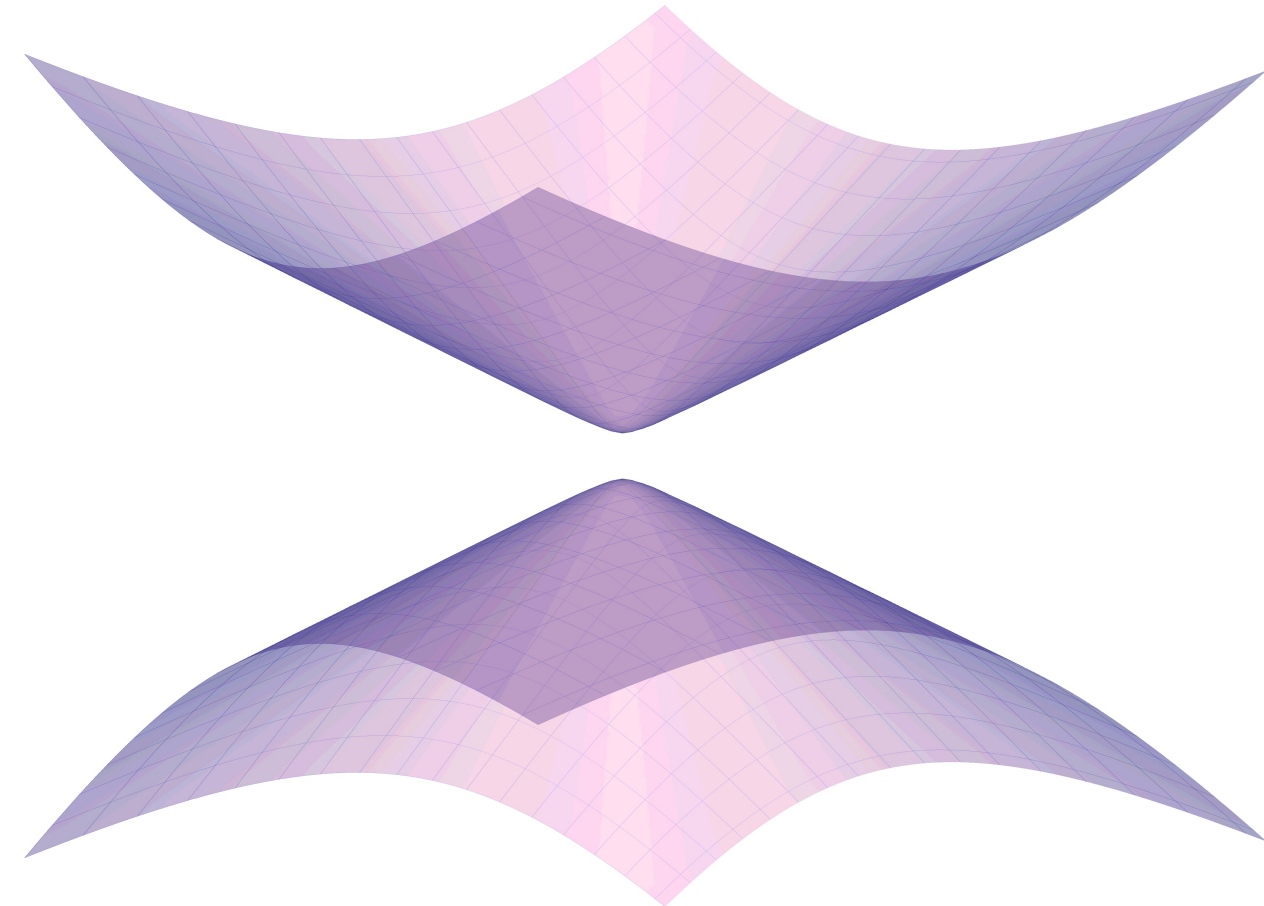
# Magnon-pair operator algebra

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**SU(2)**



**SU(1,1)**



# Quantum to semi-classical

$$\hat{\mathcal{H}} = \sum_k 2\omega_k \hat{K}_k^z + f(t) \sum_k V_k (\hat{K}_k^+ + \hat{K}_k^-)$$

Heisenberg equations

$$\begin{aligned} \frac{d\langle \hat{K}_k^z \rangle}{dt} &= if(t)V_k (\langle \hat{K}_k^- \rangle - \langle \hat{K}_k^+ \rangle) \\ \frac{d\langle \hat{K}_k^\pm \rangle}{dt} &= 2i\omega_k \langle \hat{K}_k^\pm \rangle \pm 2if(t)V_k \langle \hat{K}_k^z \rangle \end{aligned}$$

Pseudo-spin vector

$$\mathbf{J}_k = (\langle \hat{K}_k^x \rangle, \langle \hat{K}_k^y \rangle, \langle \hat{K}_k^z \rangle)$$

$$H_{\text{cl}} = \langle \hat{\mathcal{H}} \rangle = 2 \sum_k [\omega_k \mathcal{J}_k^z + f(t)V_k \mathcal{J}_k^x]$$

Dynamics on pseudo-sphere

$$\frac{d\mathbf{J}_k}{dt} = -\mathbf{J}_k \times \mathbf{B}_k$$

$$\mathbf{B}_k = -\frac{dH_{\text{cl}}}{d\mathbf{J}_k} = -2 (f(t)V_k, 0, -\omega_k)$$

$$(\mathbf{a} \times \mathbf{b})^x = a^y b^z - a^z b^y$$

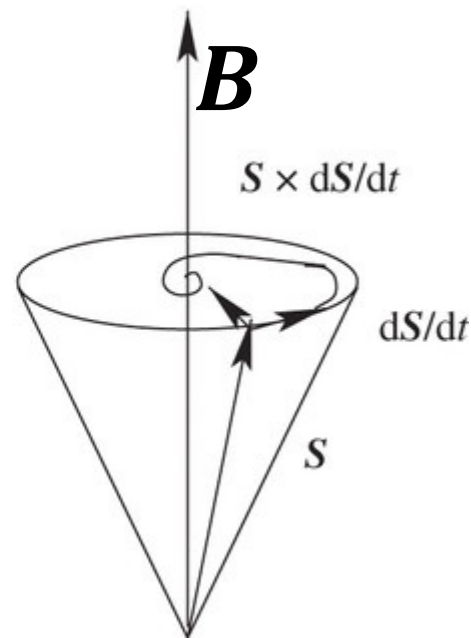
$$(\mathbf{a} \times \mathbf{b})^y = a^z b^x - a^x b^z$$

$$(\mathbf{a} \times \mathbf{b})^z = -(a^x b^y - a^y b^x)$$

# Semi-classical magnon pair dynamics

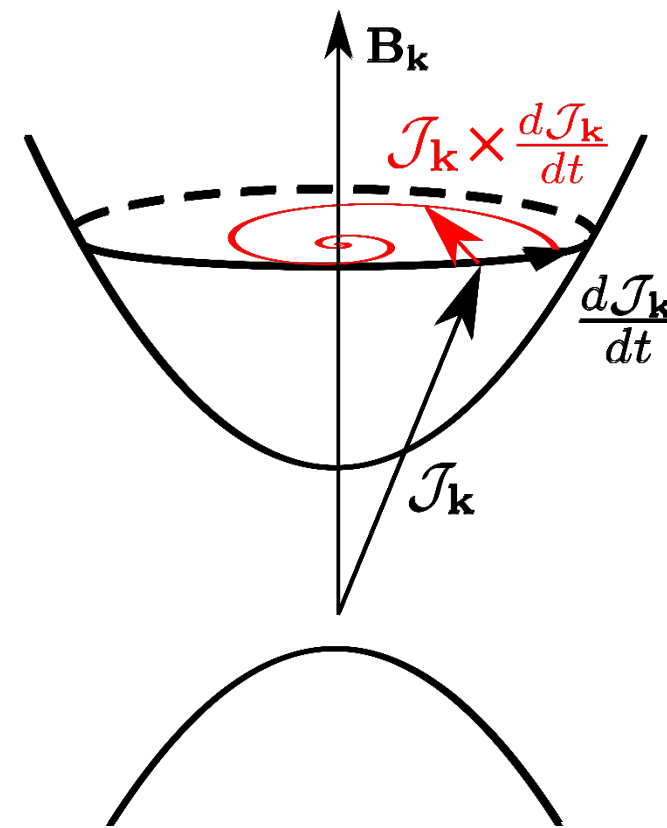
LLG equation on sphere

$$\frac{d\mathbf{S}}{dt} = -\gamma \mathbf{S} \times \mathbf{B} + \frac{\eta}{S} \mathbf{S} \times \frac{d\mathbf{S}}{dt}$$



“LLG” equation on pseudo-sphere

$$\frac{d\mathcal{J}_k}{dt} = -\mathcal{J}_k \times \mathbf{B}_k + \frac{\eta}{\mathcal{J}_k} \mathcal{J}_k \times \frac{d\mathcal{J}_k}{dt}$$



$$(\mathbf{a} \times \mathbf{b})^x = a^y b^z - a^z b^y$$

$$(\mathbf{a} \times \mathbf{b})^y = a^z b^x - a^x b^z$$

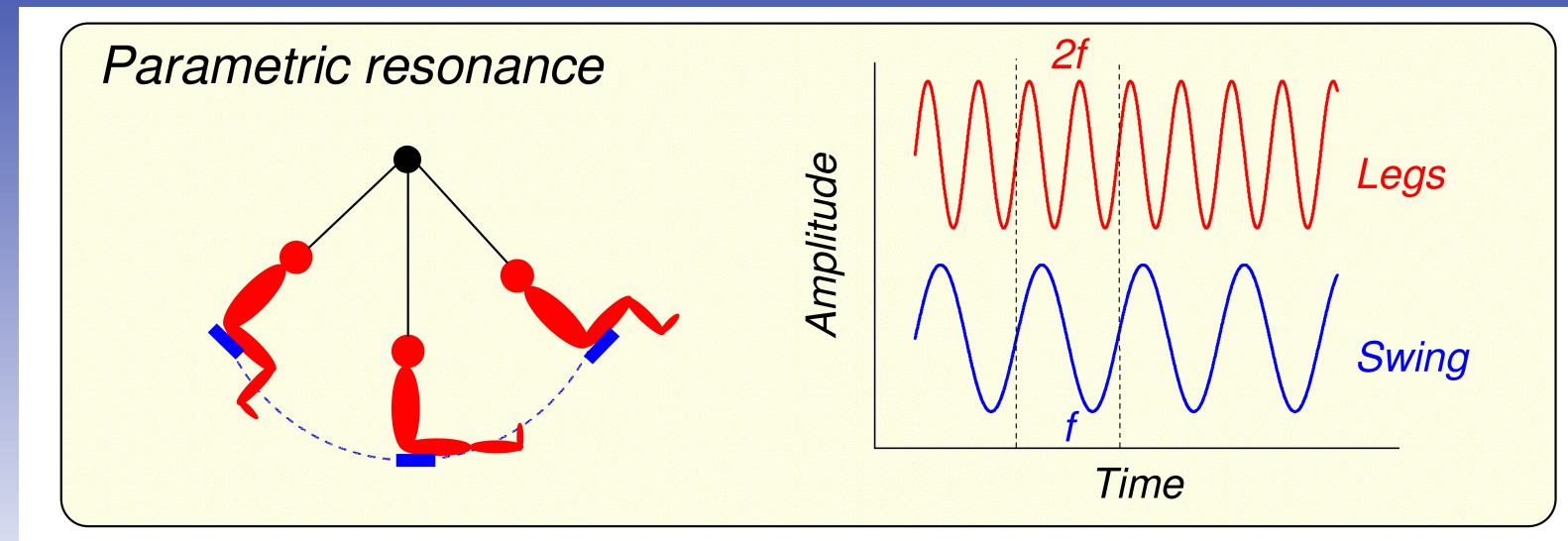
$$(\mathbf{a} \times \mathbf{b})^z = a^x b^y - a^y b^x$$

$$(\mathbf{a} \times \mathbf{b})^x = a^y b^z - a^z b^y$$

$$(\mathbf{a} \times \mathbf{b})^y = a^z b^x - a^x b^z$$

$$(\mathbf{a} \times \mathbf{b})^z = -(a^x b^y - a^y b^x)$$

# Parametric oscillations



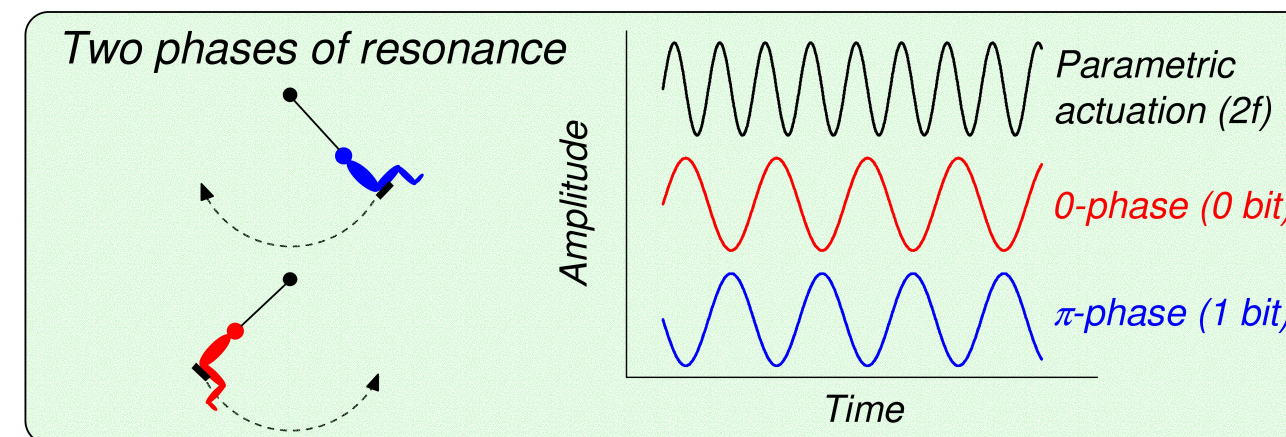
$$\left[ \frac{d^2}{dt^2} + \eta\omega_0 \frac{d}{dt} + \omega_0^2(1 + \alpha + \beta x^2 - 2\Gamma \sin 2\omega_0 t) \right] x(t) = 0$$

Ansatz:  $(\alpha = \beta = 0)$

$$x(t) = x_c(t) \cos \omega_0 t + x_s(t) \sin \omega_0 t$$

$$x_s = x_s(0) e^{\omega_0(\Gamma - \eta)t}$$

$$x_c = x_c(0) e^{-\omega_0(\Gamma + \eta)t}$$



# Parametric excitation of magnon pairs

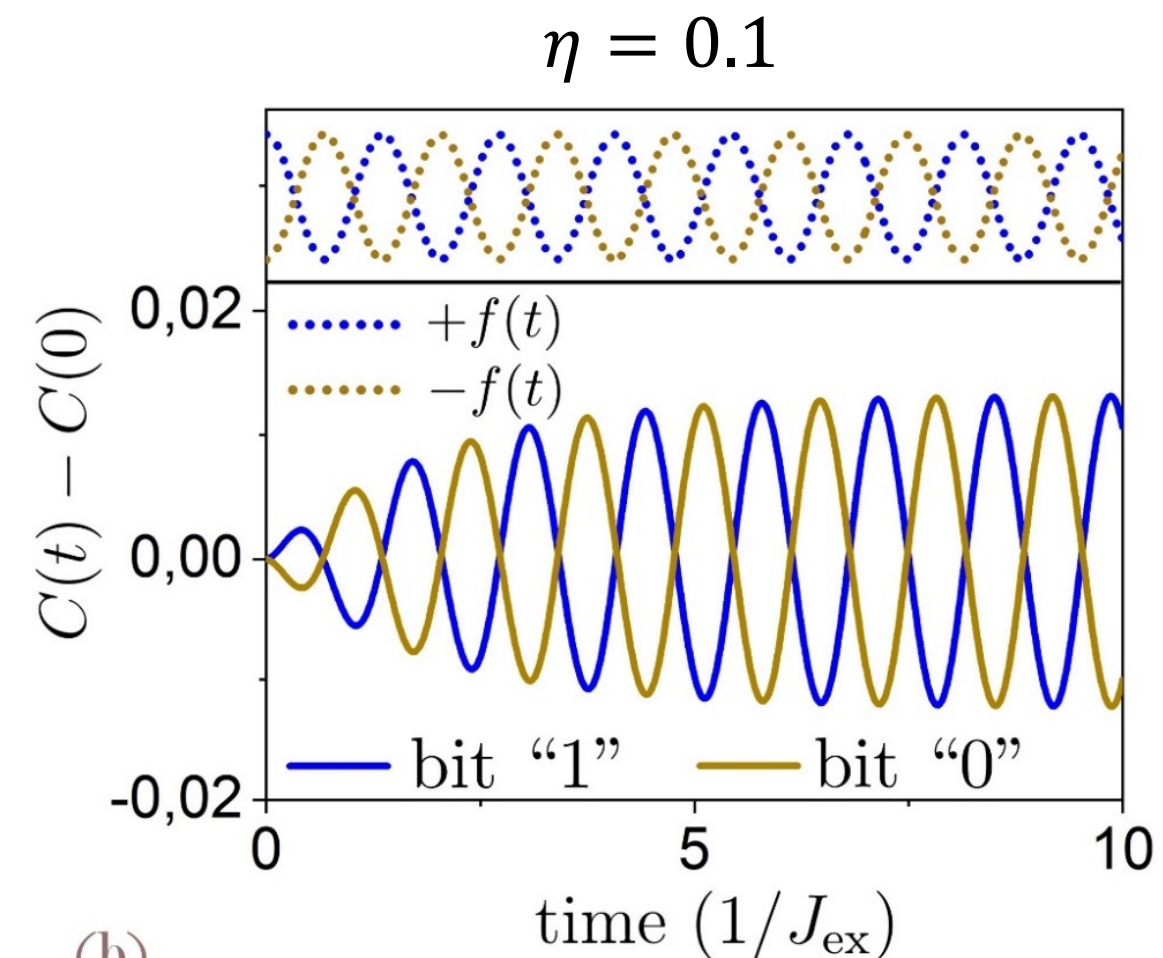
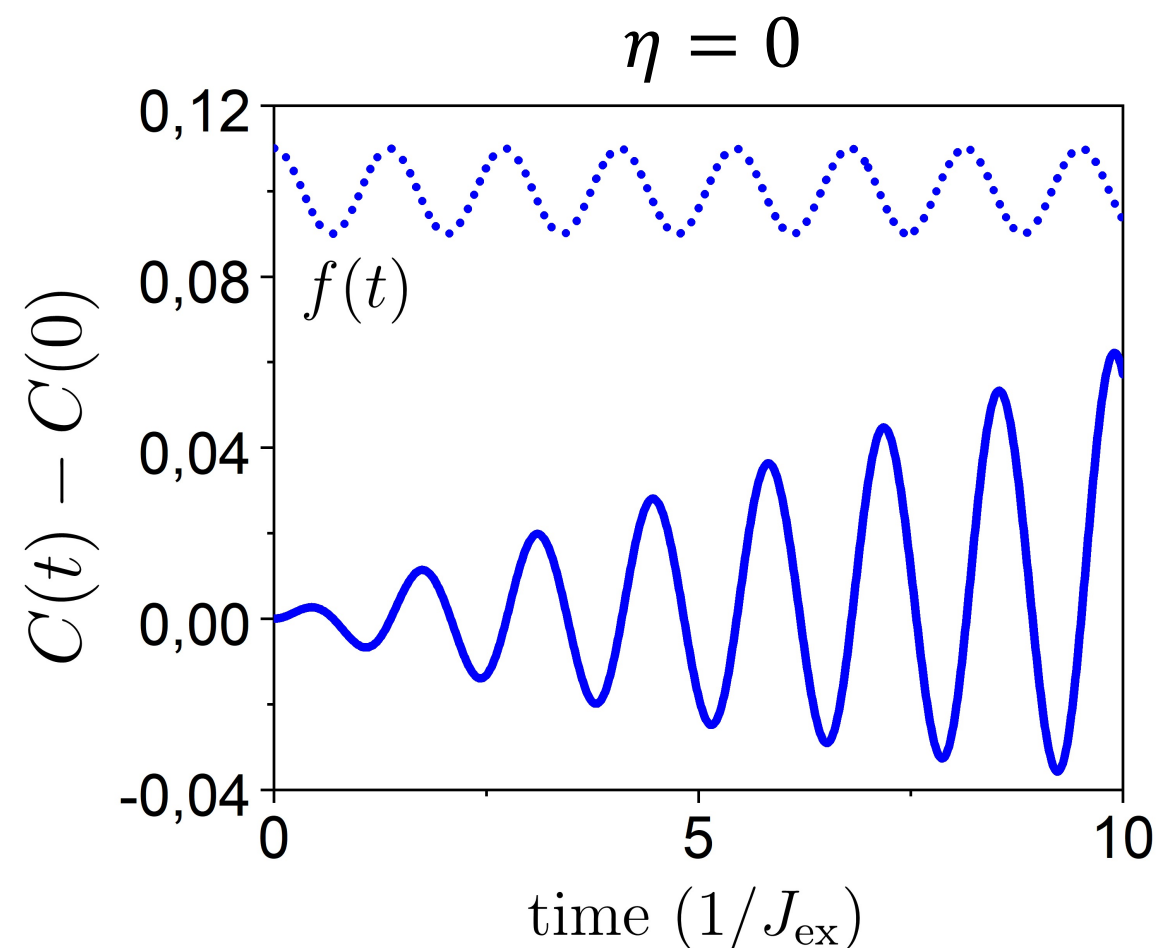
Excitation of parametric resonance by periodic modulation  $f(t) = \cos(2\omega_M t)$

$$\frac{d\mathcal{J}_k}{dt} = -\mathcal{J}_k \times \mathbf{B}_k + \frac{\eta}{J_k} \mathcal{J}_k \times \frac{d\mathcal{J}_k}{dt}$$

$$\mathbf{B}_k = -2(V_k \cos(2\omega_M t), 0, -\omega_k)$$

$$C(t) = \langle \hat{\mathbf{S}}_i \cdot \hat{\mathbf{S}}_{i+\delta x} \rangle = \sum_k \Theta_k \cdot \mathcal{J}_k$$

Bits as oscillation states

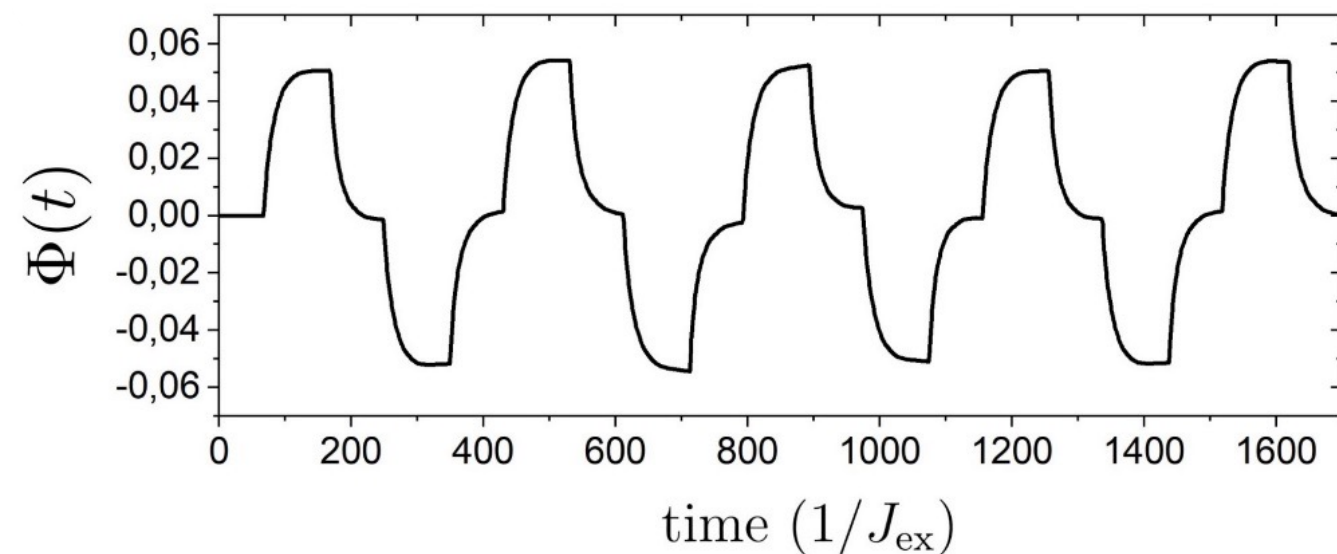
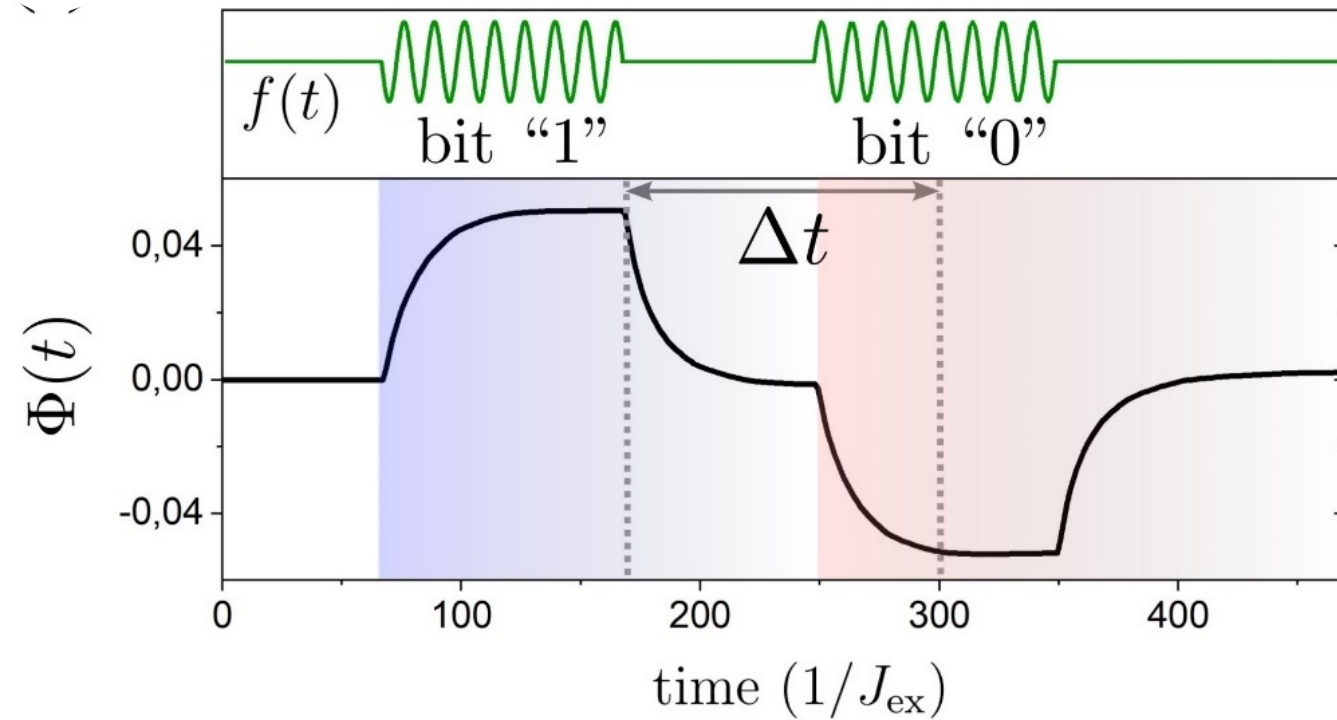




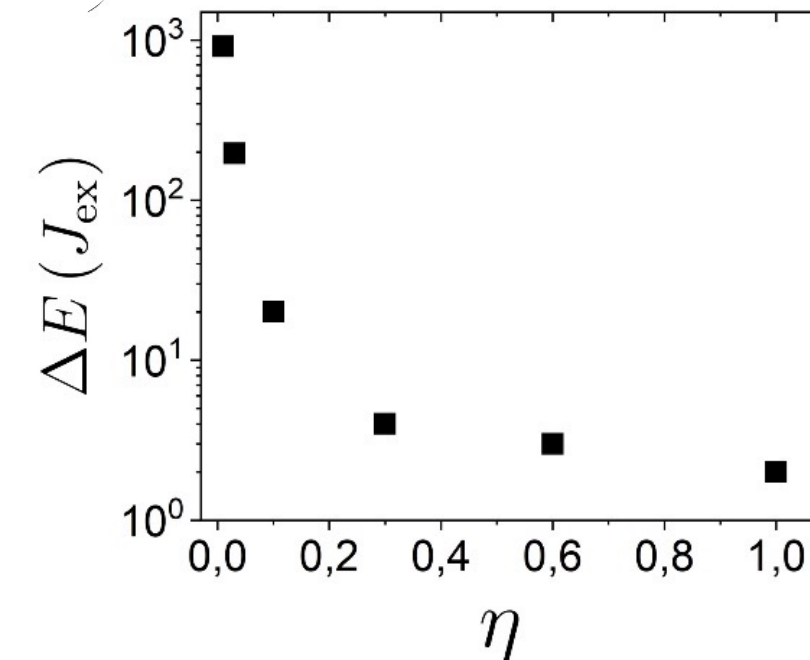
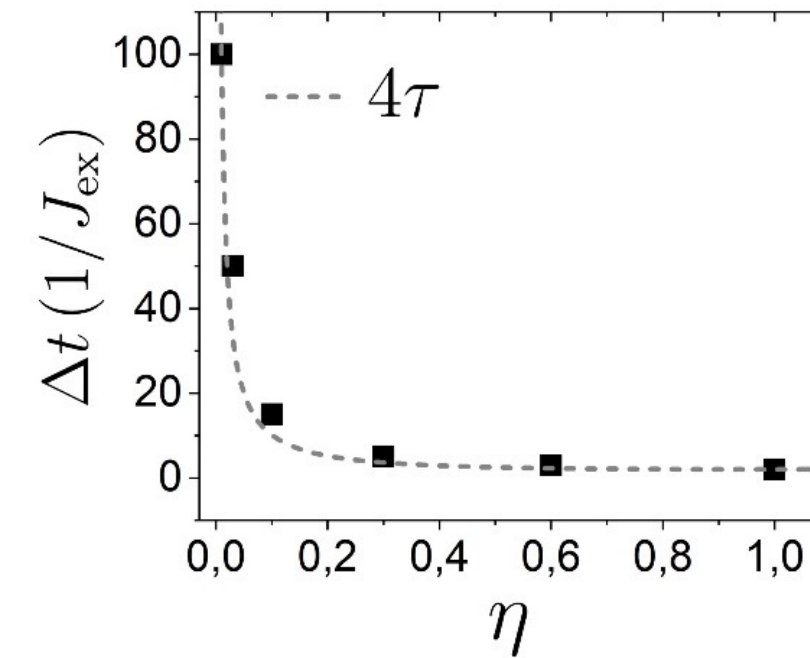
# Bits as phases of magnon pair oscillations

$$\Phi(t) = \int_t^{t+T} dt' C(t') \cdot \cos 2\omega_k t'$$

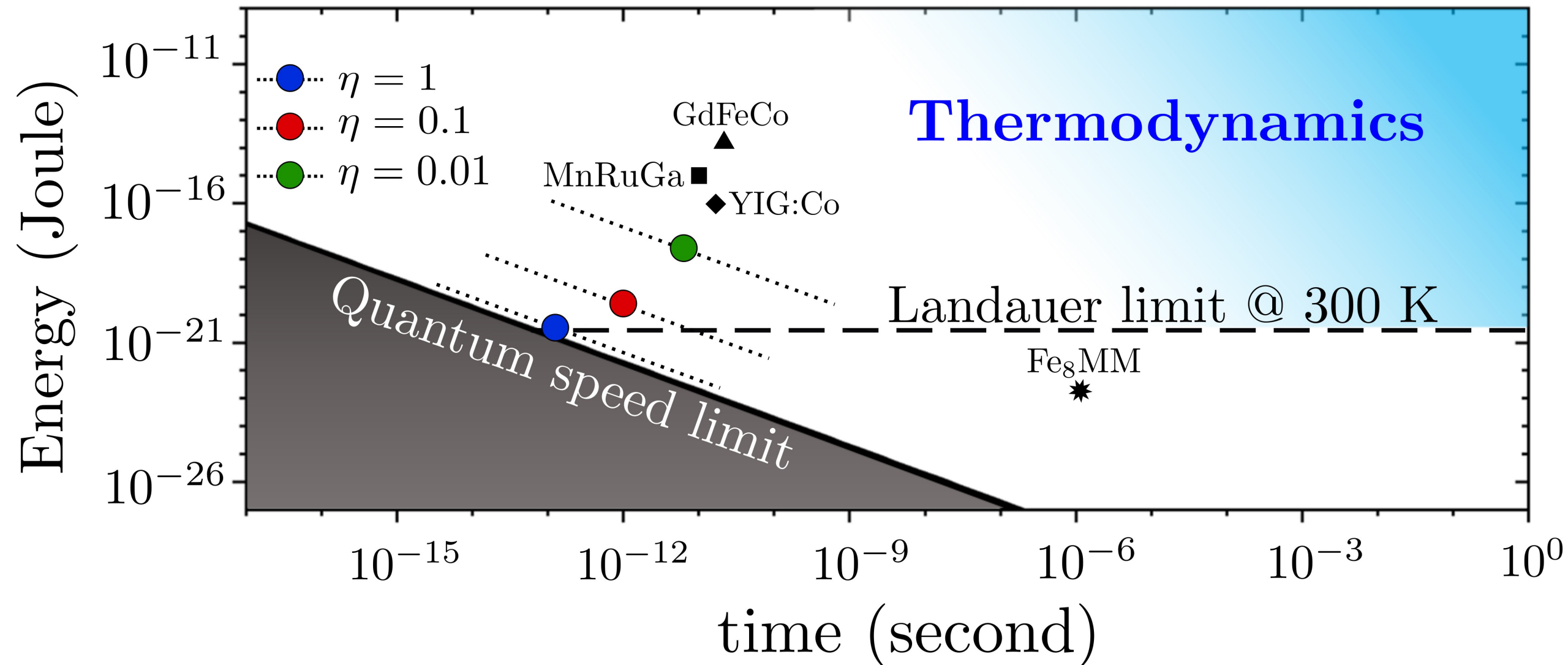
$\eta = 0.01$



$$J_{ex} = 10^{-2} eV \Rightarrow t = 100/J_{ex} \sim 6 \text{ ps}$$



# Approaching the fundamental limits



- Strong damping  $\eta \sim 1$  approaches “ultimate” physical limits
- Up to 5 orders of magnitude better than best magnets so far

**GdFeCo:** K. Vahaplar Phys. Rev. Lett. 103, 117201(2009)

**MnRuGa:** C. Banerjee, et al., Nature communications 11, 4444 (2020)

**YIG:Co:** A. Stupakiewicz, Nature 542, 71–74 (2017).

**$\text{Fe}_8\text{MM}$ :** R. Gaudenzi, et al., Nature Phys. 14,565–568 (2018)

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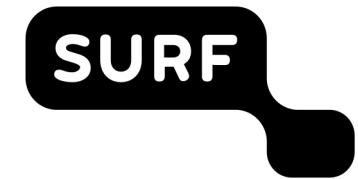
*(now Konstanz, Germany)*

D. Bossini

*SPICE/ Uni Mainz, Germany*

E.V. Gomonay

Institute for  
Molecules and Materials  
Radboud University



Openings for PhD/PD, to be announced online soon!

[j.mentink@science.ru.nl](mailto:j.mentink@science.ru.nl)



# Summary

- **Supermagnonic propagation**  
magnons at the edge can propagate up to 40% faster  
extraordinary strong magnon-magnon interactions in 2D
- **Parametric excitation of magnon-pairs**  
Semi-classical dynamics reveals switching near fundamental limits

