INSTITUTE FOR QUANTUM MATTER

A collaboration between
JOHNS HOPKINS UNIVERSITY
and PRINCETON UNIVERSITY

Neutron Scattering Experiments for Quantum and Frustrated Spin Chains

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Previously at:

Institut Laue Langevin, Grenoble, France

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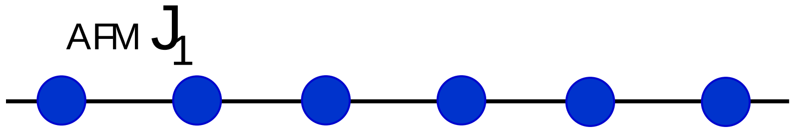
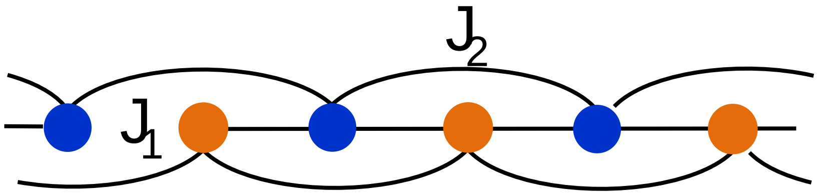
JOHNS HOPKINS
UNIVERSITY

Outline

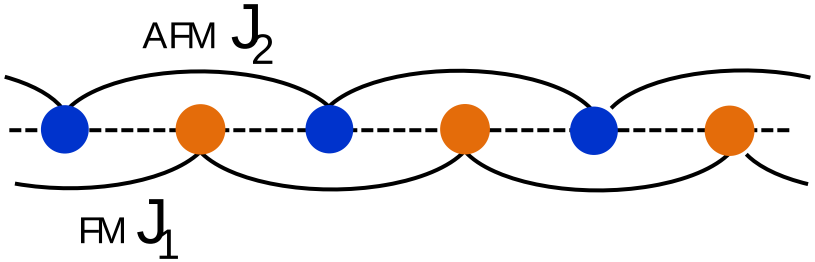
<u>Framework:</u> (Quasi-) 1D Heisenberg quantum (S=1/2) spin chains

1. Introduction



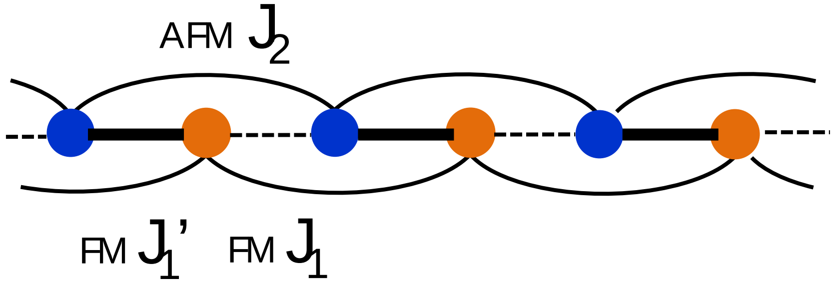


2. Frustrated ferromagnetic chains in $LiCuVO_4$



M. Enderle *et al.*, PRL **104**, 237207 (2011)
M. Mourigal, *et al.*, PRB **83**, 100409(R) (2011)
M. Mourigal, *et al.*, PRL **109**, 027203 (2012)

3. Frustrated alternating ferromagnetic chains in $LiCuSbO_4$



S. Dutton *et al.*, PRL **108**, 187206 (2012)
M. Mourigal *et al.*, *work in progress*

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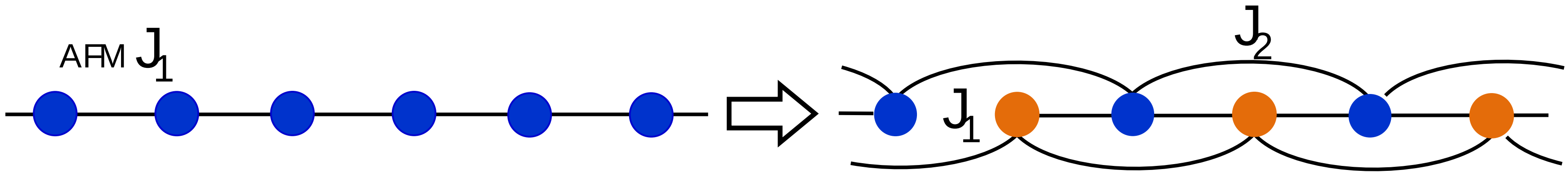
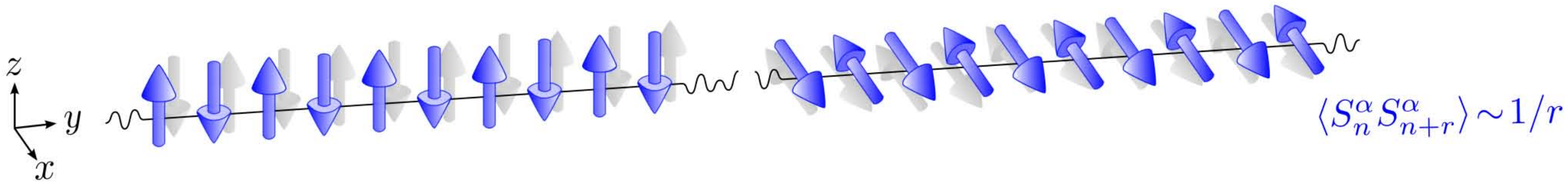


1.1 Introduction

The spin-1/2 Heisenberg *n.n.* chain is well understood

❖ Ground-state



Algebraic spin correlations, no long-range-order, singlet with macroscopic entanglement

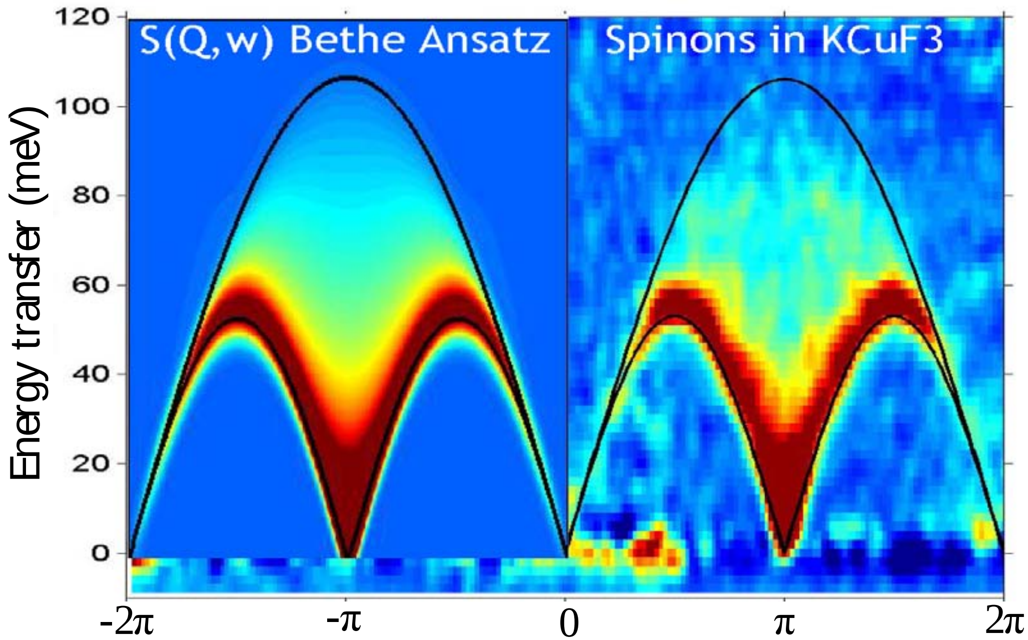
1.1 Introduction

The spin-1/2 Heisenberg *n.n.* chain is well understood

❖ Excitations Dynamical correlations are known exactly for T=0!

Caux *et al.*,
J. Stat. Mech. '06

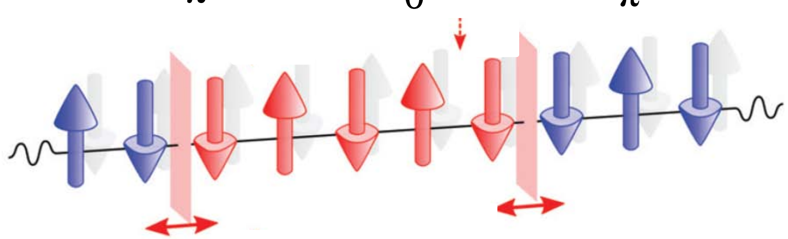
Tennant, Lake *et al.*
Nature Materials '05



2-spinons (72.89%)

4-spinons (26%)

6-spinons (...)

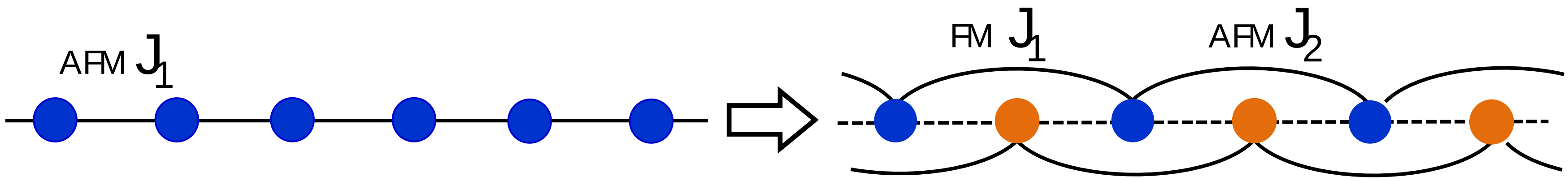


From J.S Caux website

Courtesy of A. Tennant

1.2 Introduction

What is the result of adding frustration via AFM *n.n.n* exchange?



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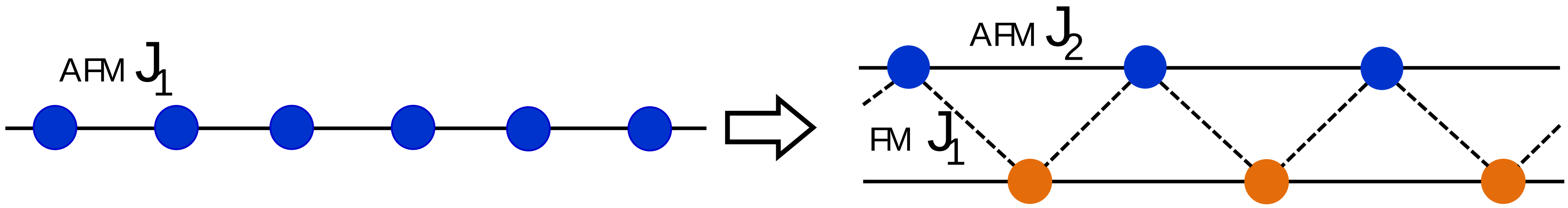
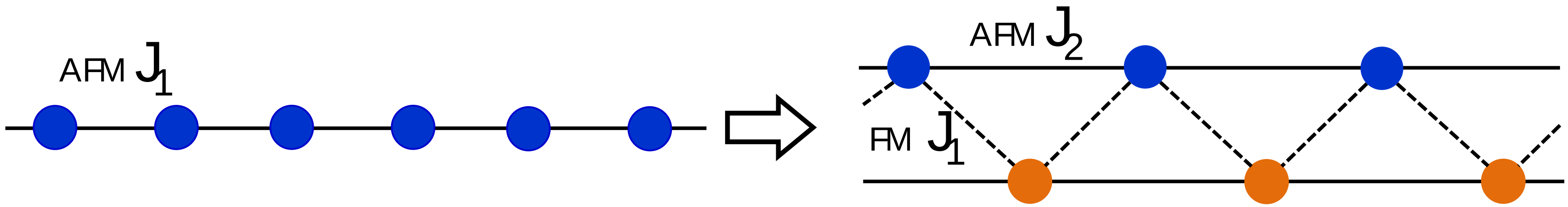


General Hamiltonian $$\mathcal{H} = \sum_i \{J_1(\mathbf{S}_i \cdot \mathbf{S}_{i+1})_\Delta + J_2(\mathbf{S}_i \cdot \mathbf{S}_{i+2})_\Delta - hS_i^z\}$$

Quantum spins $S = 1/2$ Easy-plane anisotropy D

1.2 Introduction

What is the result of adding frustration via AFM *n.n.n* exchange?



❖ Variety of exotic spin-nematic ground-states

Vector-Chiral

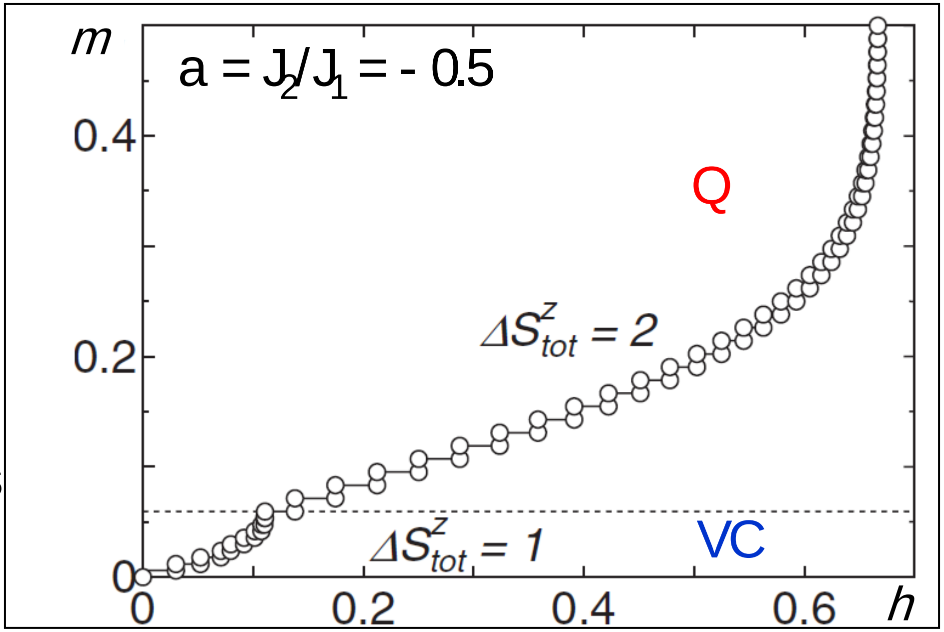
$$\boldsymbol{\kappa}_i^n = (\mathbf{S}_i \times \mathbf{S}_{i+n})^z$$

Quadrupolar

$$\mathrm{Q}_{x^2-y^2} - i\mathrm{Q}_{xy} = S_i^- S_j^-$$

Bose-Einstein condensation of magnon pairs

DMRG



See: Chubukov, PRB '91 | Vekua *et al.*, PRB '07 | **Hikihara *et al.*, PRB '08** | Sudan *et al.*, PRB '08

1.3 Introduction

Where to find candidate materials ?

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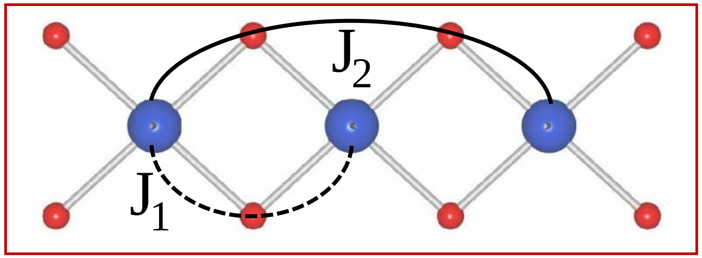
❖ Edge-sharing copper-oxide chains



Jahn-Teller distorted CuO_6 octahedral

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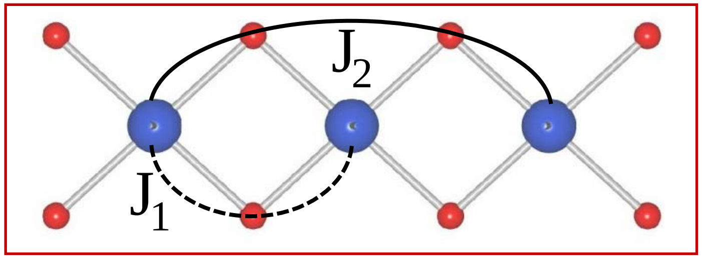

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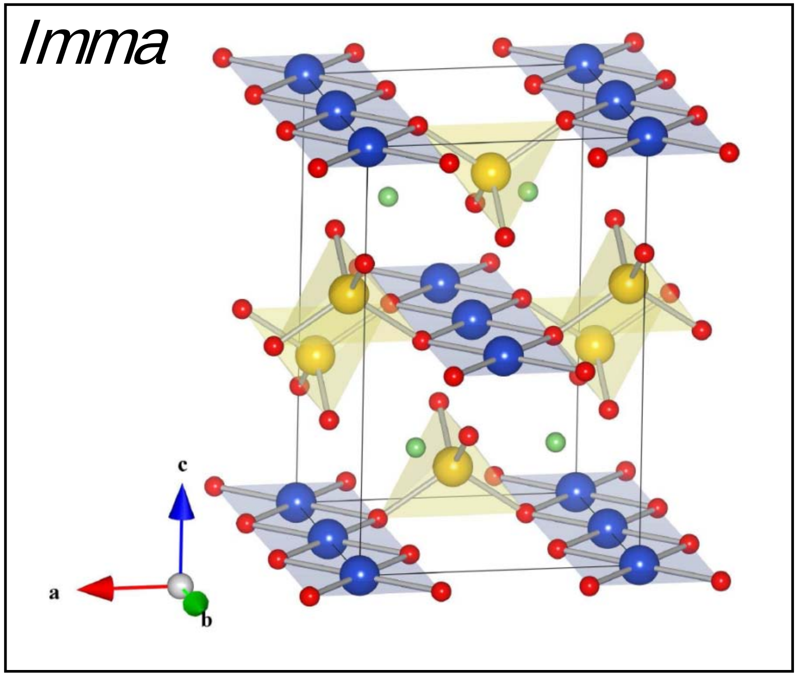
$LiCuVO_4$



See: La Fontaine *et al.*, Acta Cryst. '89 | **H.-J. Koo *et al.* Inorg. Chem. '11** | Dutton *et al.* PRL '12

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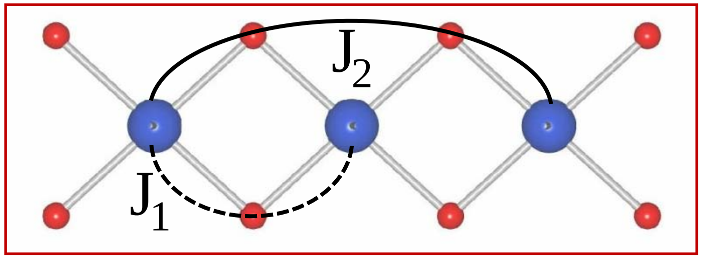

Jahn-Teller distorted CuO_6 octahedral

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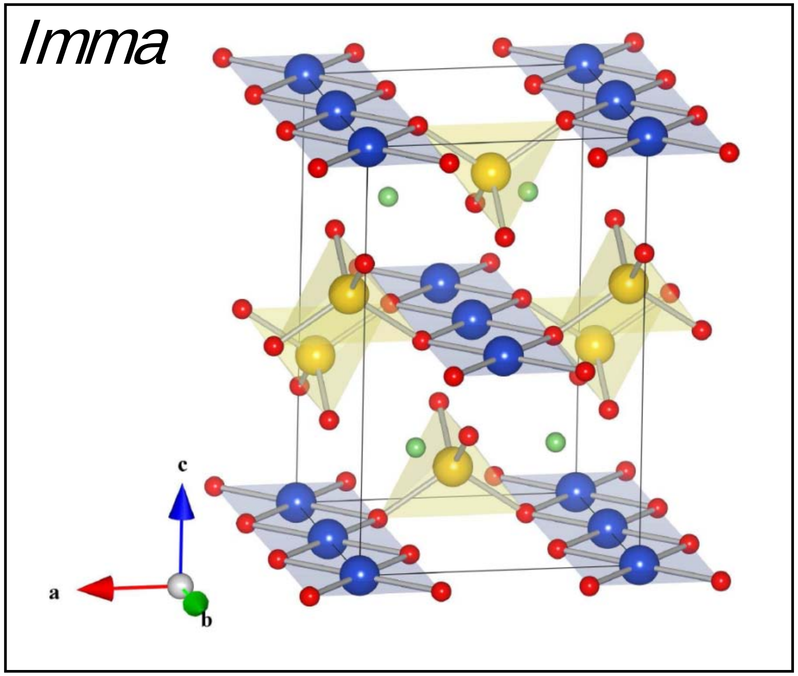
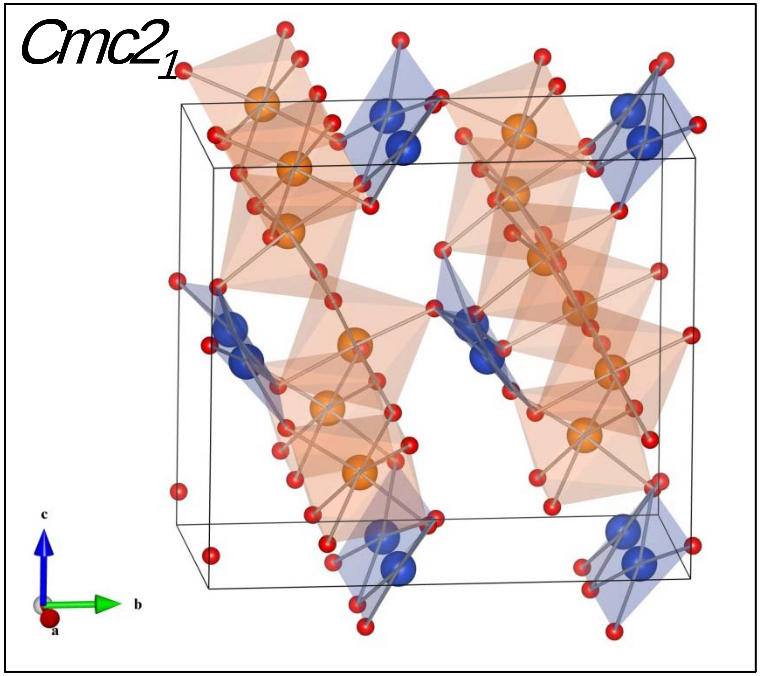


$LiCuSbO_4$



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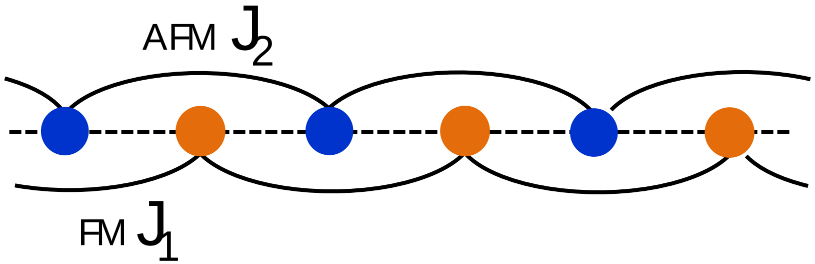

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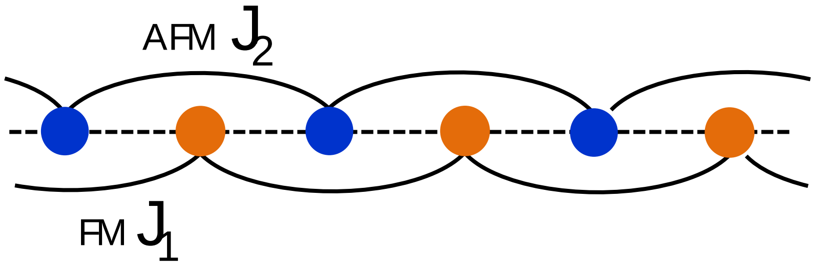

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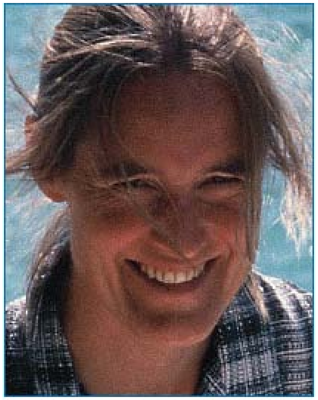
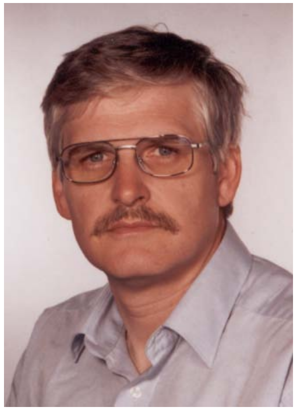


M. Enderle
(ILL, Grenoble)

B. Fåk
(CEA, Grenoble)



R. Kremer
(Max-Planck, Stuttgart)

J. M. Law (Max-Planck, Stuttgart)

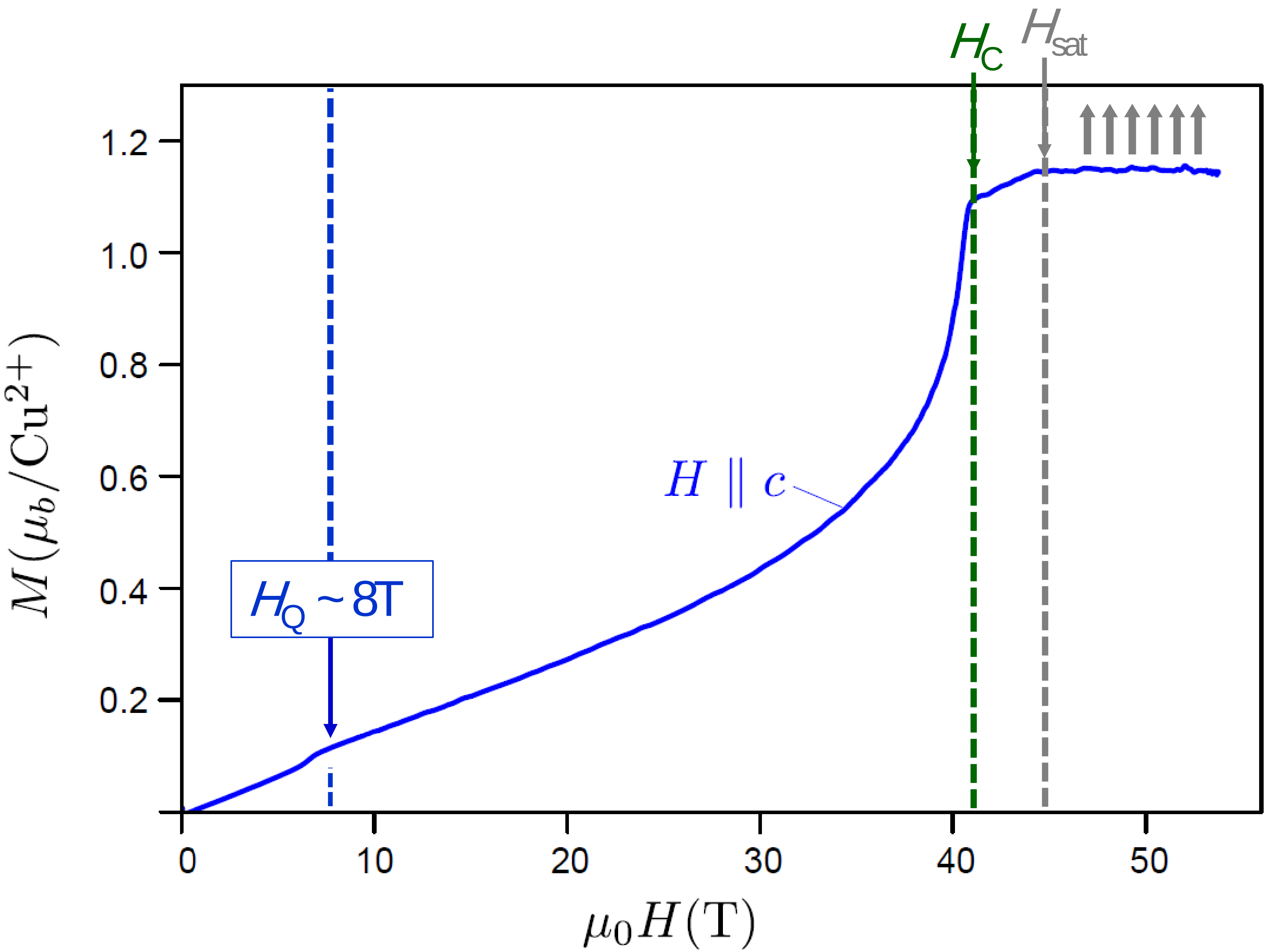
A. Prokoviev (Vienna)

A. Schneidewind (Munich)

A. Hiess (ESS, Lund)

M. Zhitomirsky (Grenoble)

2.1 The paradigmatic material $LiCuVO_4$



See: Svistov *et al.*, JETP Letters '11

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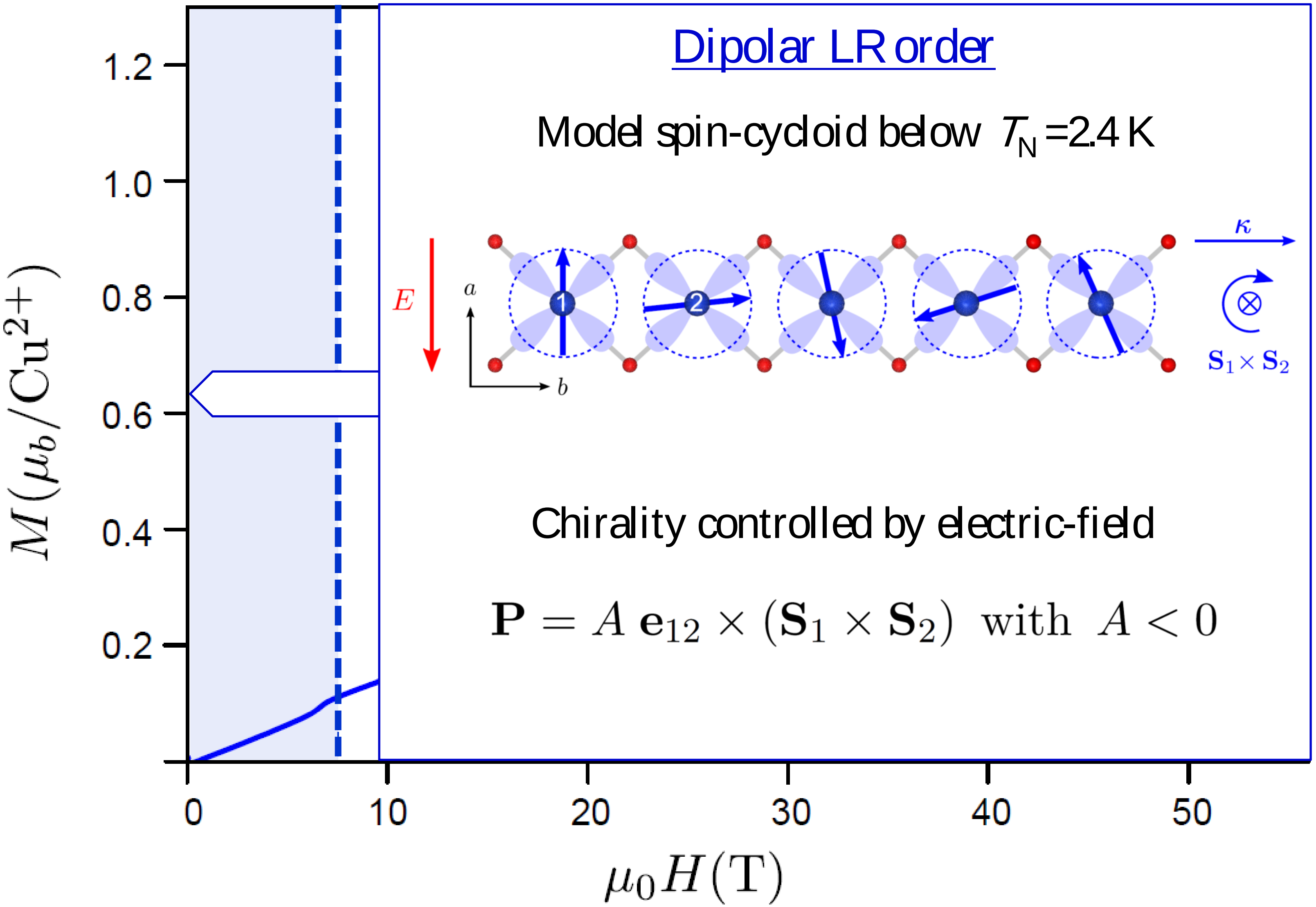

See: Gibson *et al.*, Physica B '04 | **Mourigal *et al.* PRB '11** |

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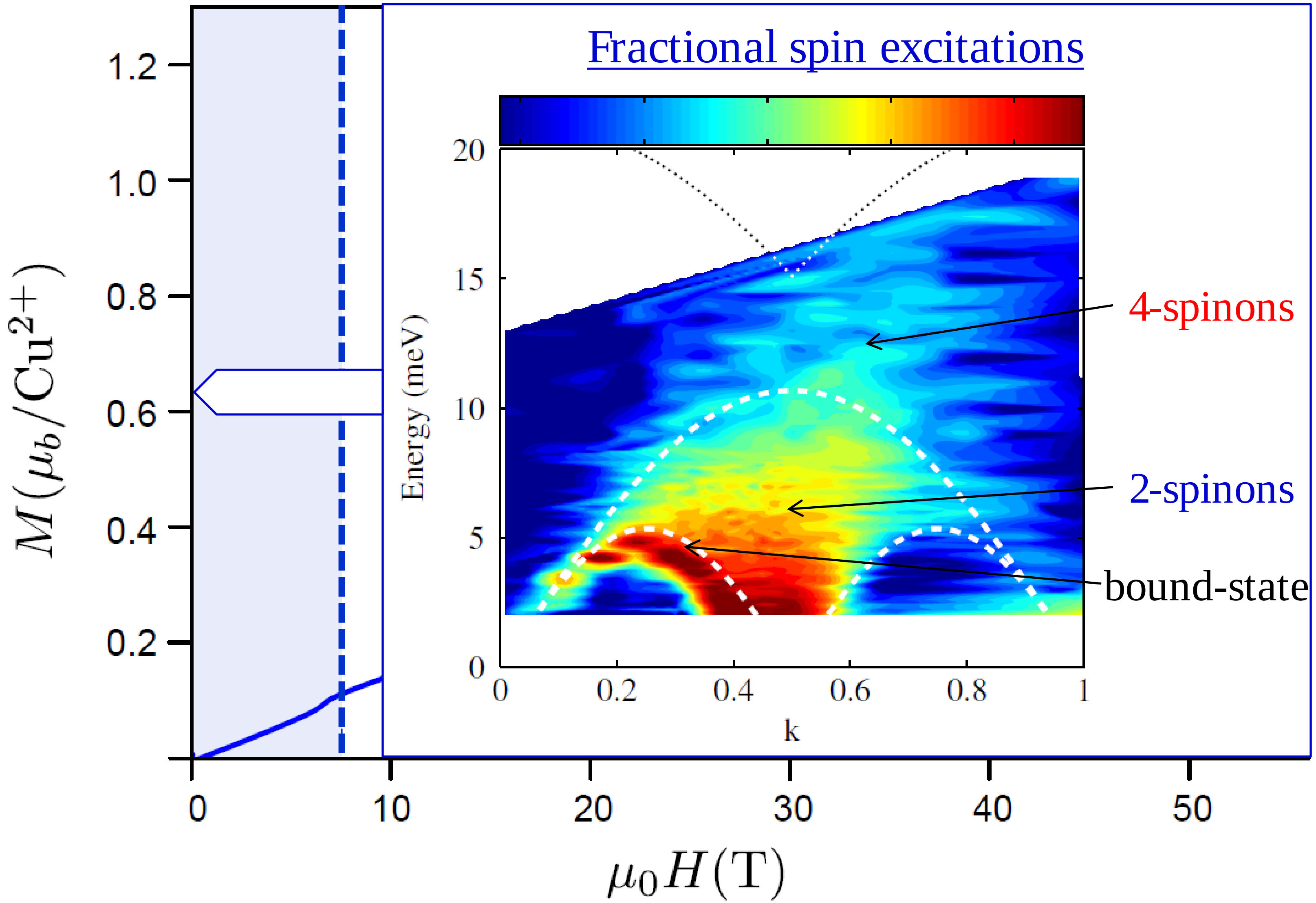

See: Gibson *et al.*, Physica B'04 | Mourigal *et al.* PRB'11 | **Enderle *et al.* PRL '10**

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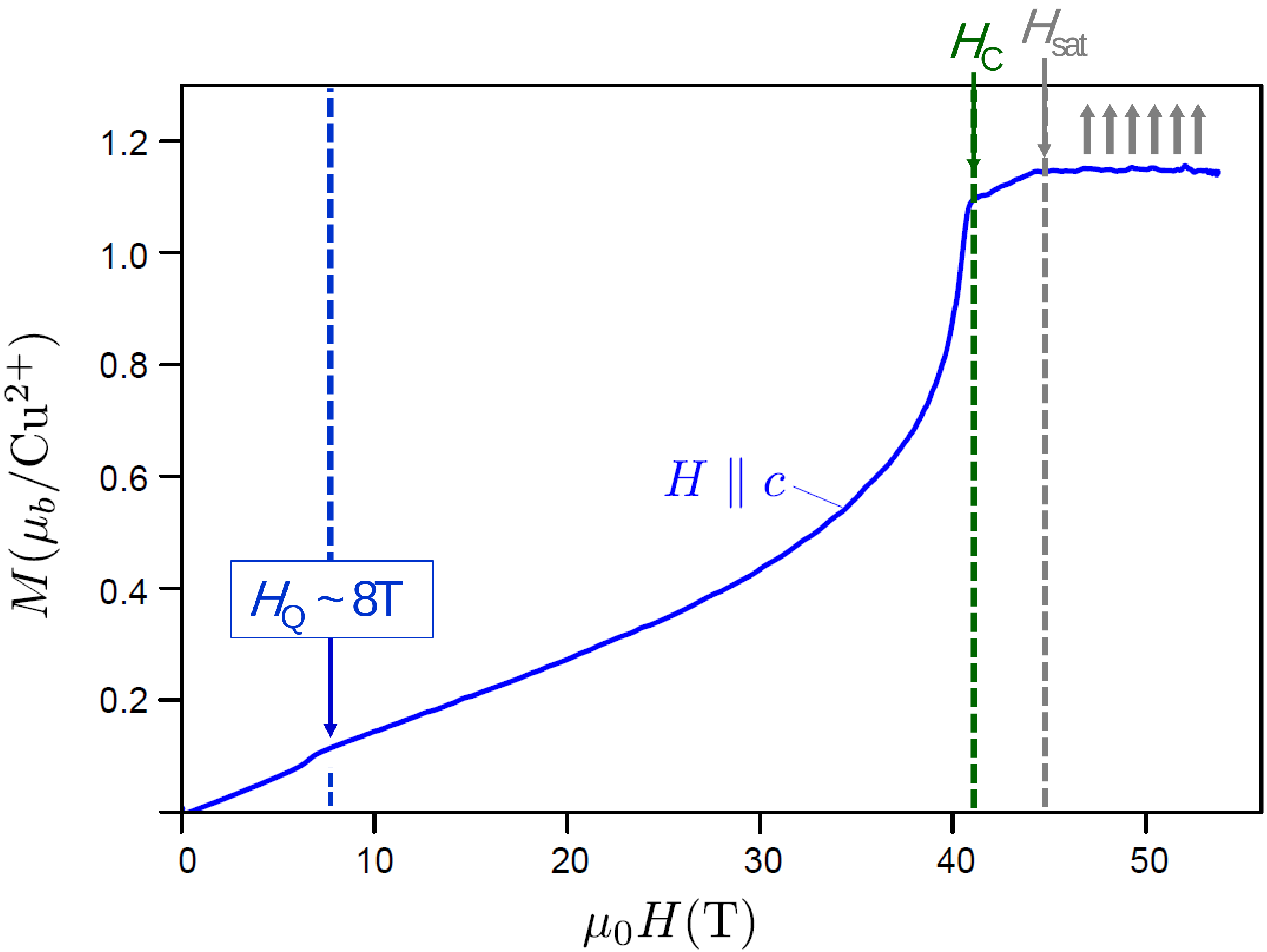

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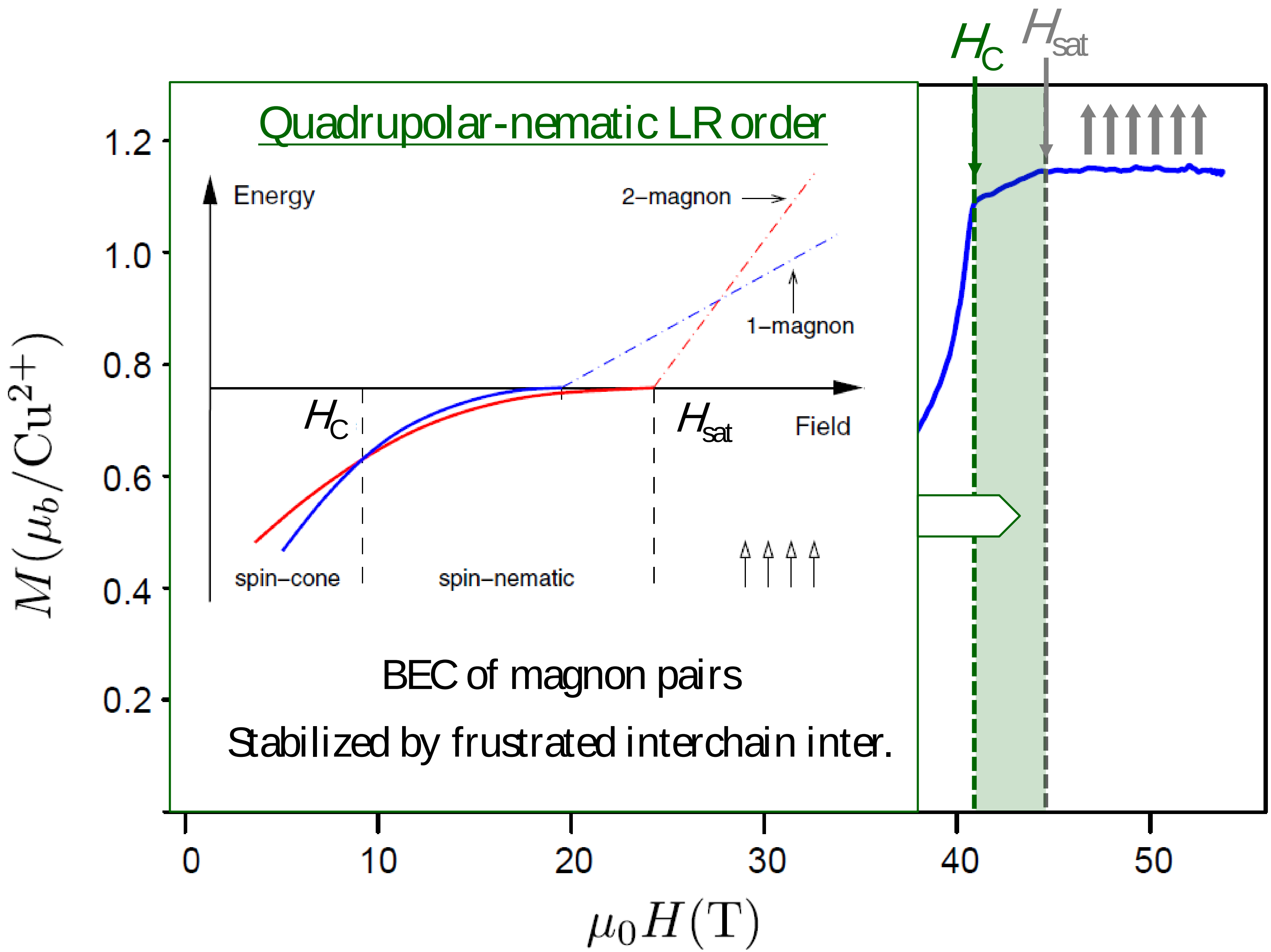

See: Svistov *et al.*, JETP Letters'11 | **Zhitomirsky and Tsunetsugu, EPL '10**

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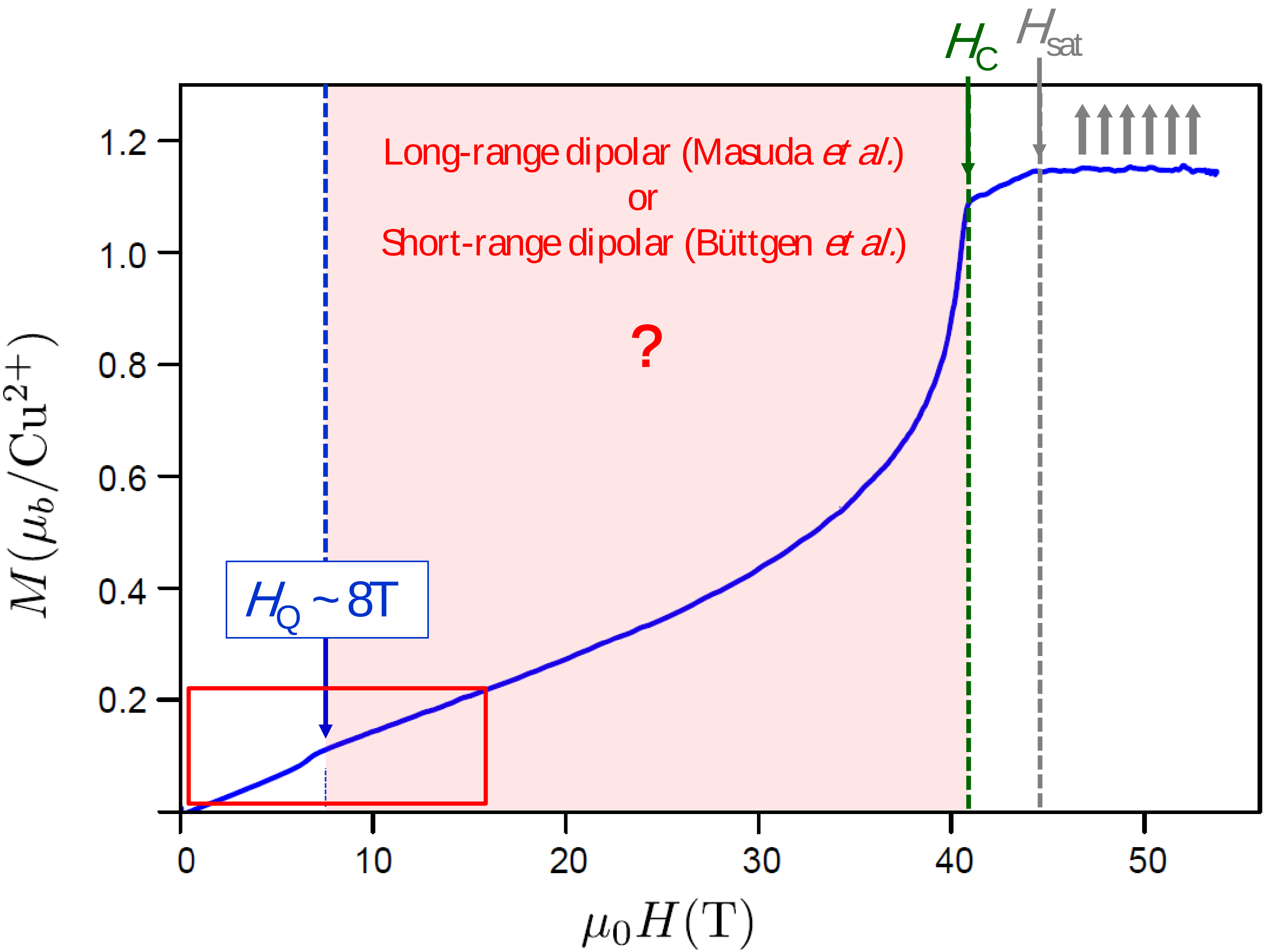
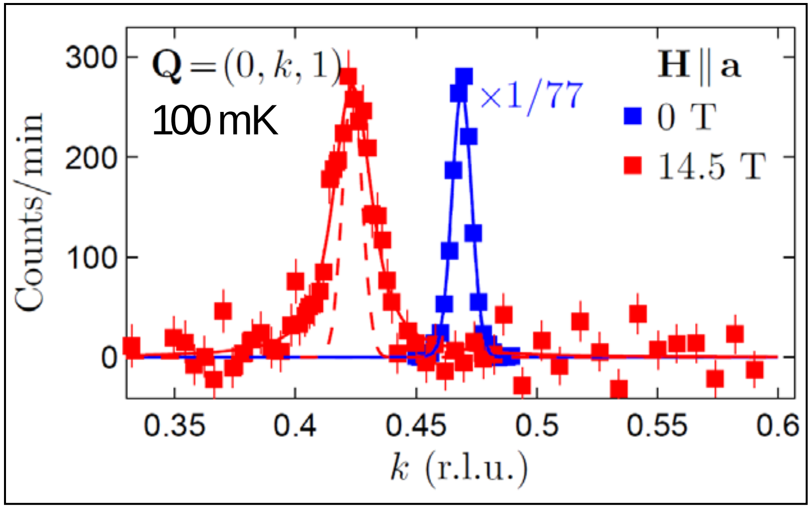
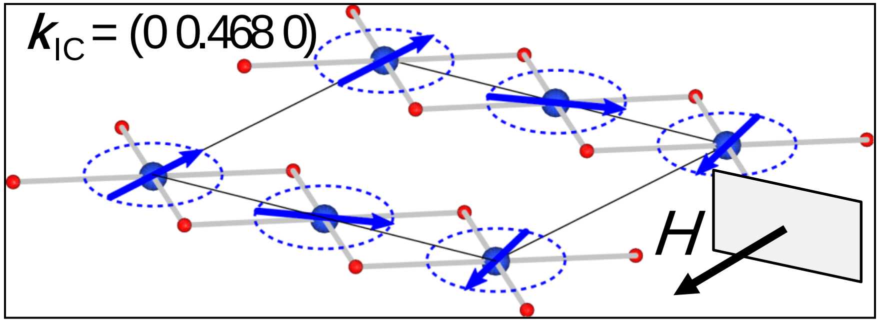


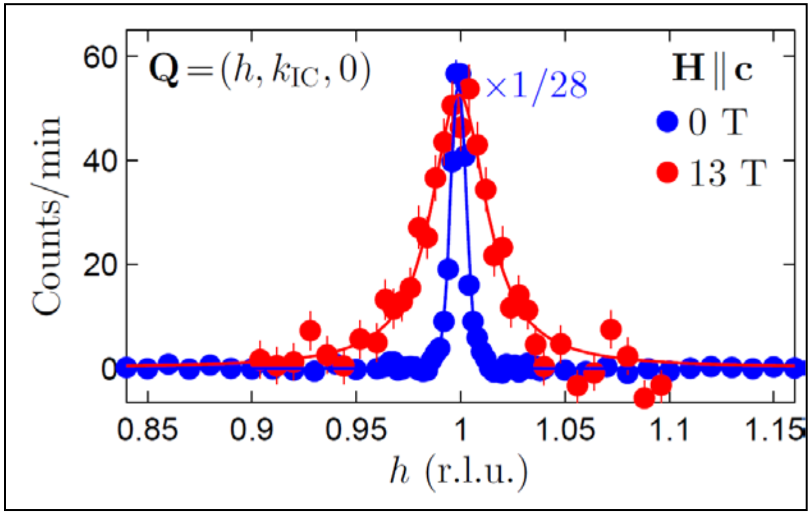
See: Svistov *et al.*, JETP Letters'11 | Buttgen *et al.* PRB'07; PRB'10, PRB' 12 | Masuda *et al.* JPSJ'11

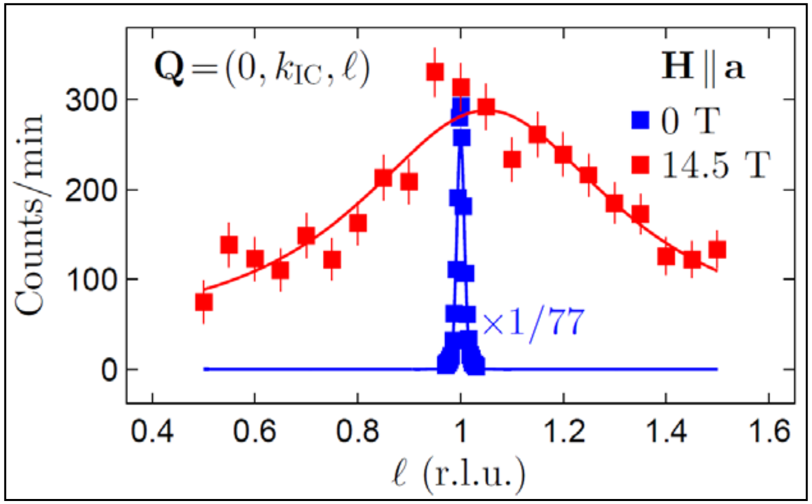
2.2 Elastic neutron scattering above H_Q

❖ 1. Dipolar spin correlations become short-ranged









Instruments: PANDA, FRM-II, Munich and IN14, ILL, Grenoble

2.2 Elastic neutron scattering above H_Q

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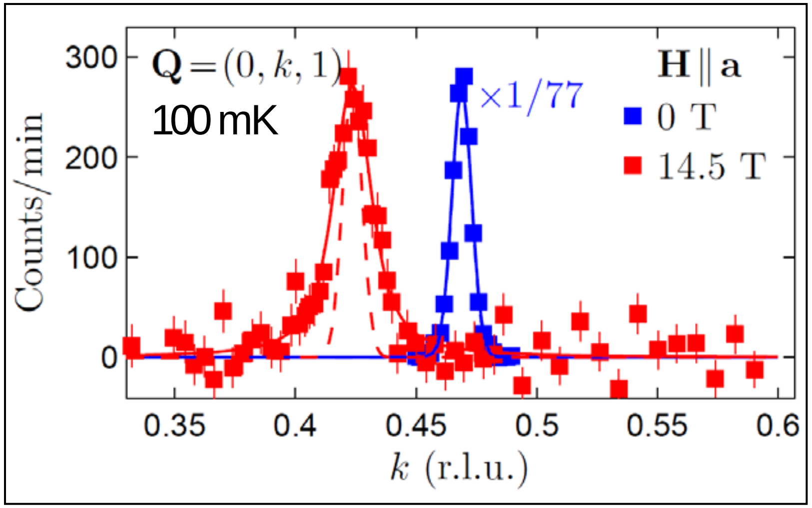
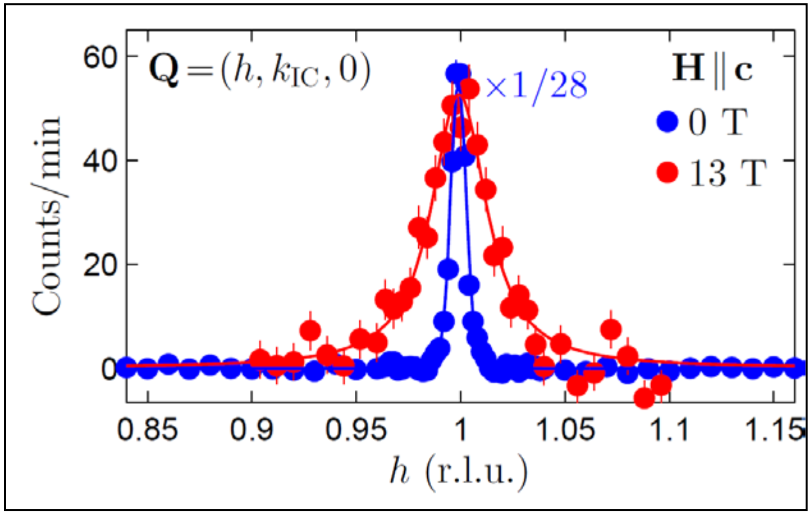
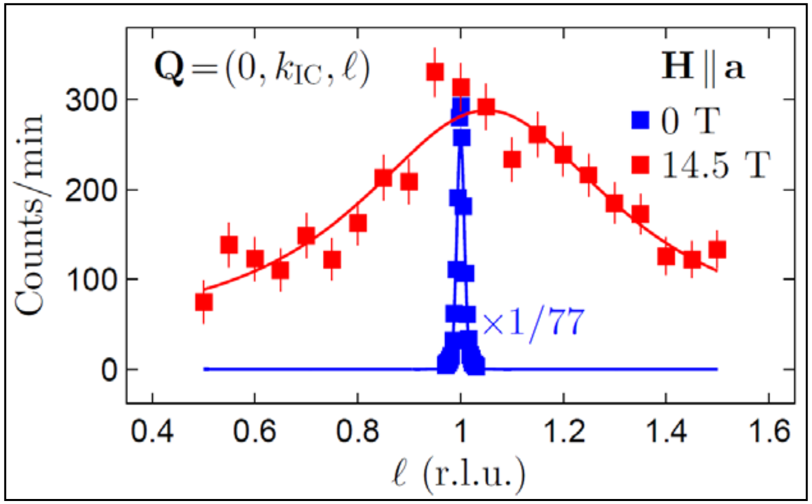


Dipolar correlations are short-range in all directions above H_Q at 100 mK

Integrated intensity is conserved





Instruments: PANDA, FRM-II, Munich and IN14, ILL, Grenoble

2.2 Elastic neutron scattering above H_Q

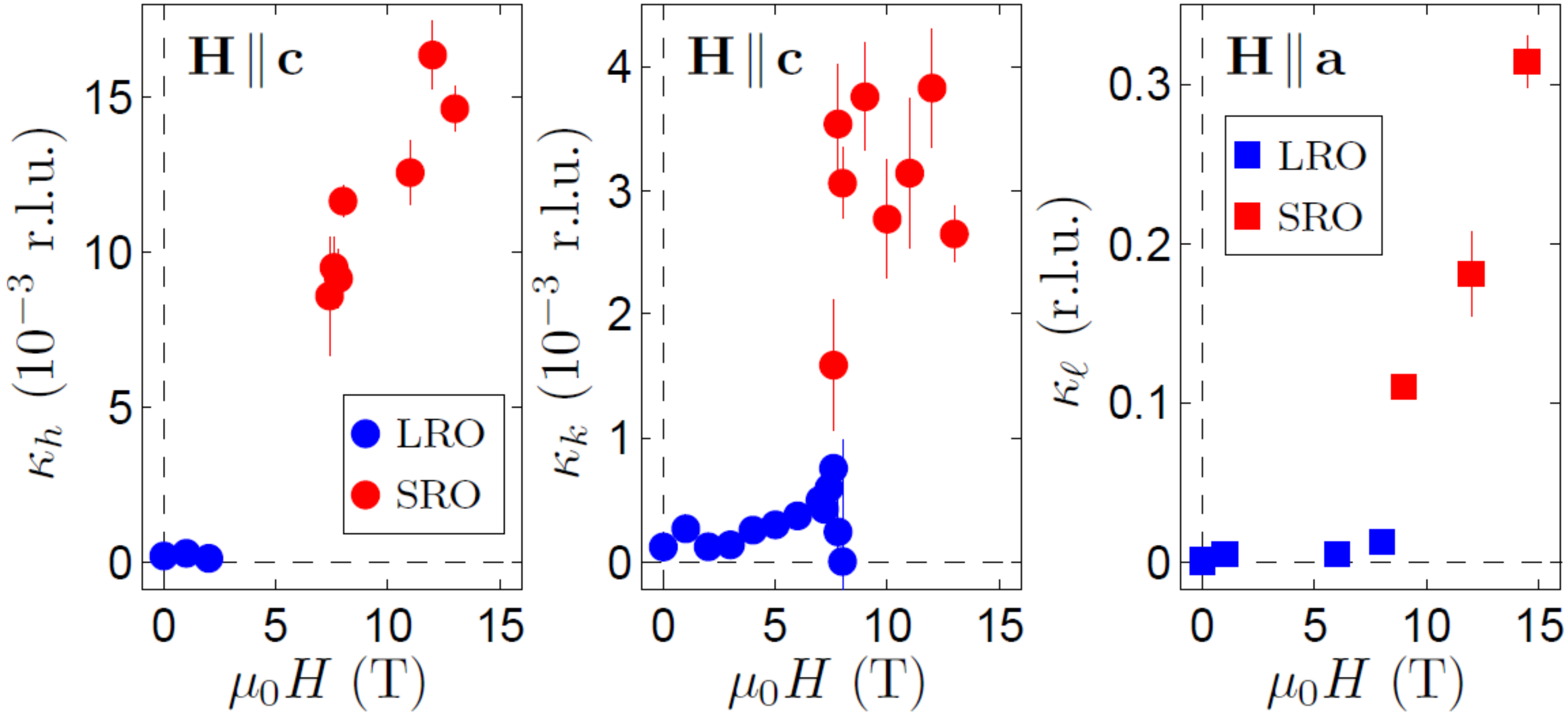
❖ 1. Dipolar spin correlations become short-ranged

Abrupt broadening at H_Q

Dipolar correlations are short-range in all directions above H_Q at 100 mK

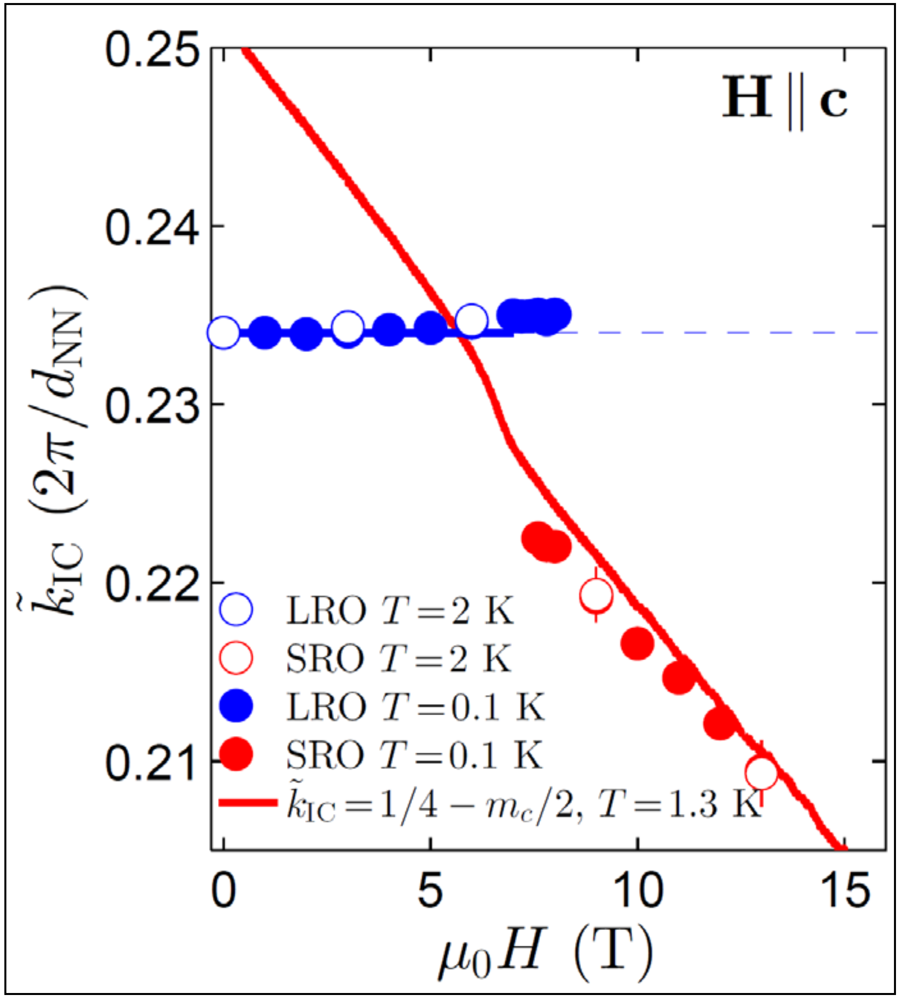
x_a ~70 nn, x_b ~700 nn, x_c ~6 nn

Integrated intensity is conserved

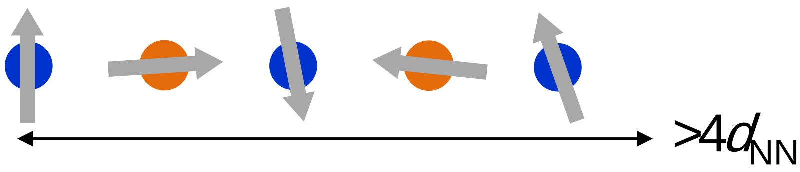


2.2 Elastic neutron scattering above H_Q

❖ 2. Field-dependence of short-range dipolar correlations



LR Dipolar order



$$\boxed{\tilde{k}_{IC}=1/4-\delta}$$

SR Dipolar order

In the quadrupolar-nematic phase:

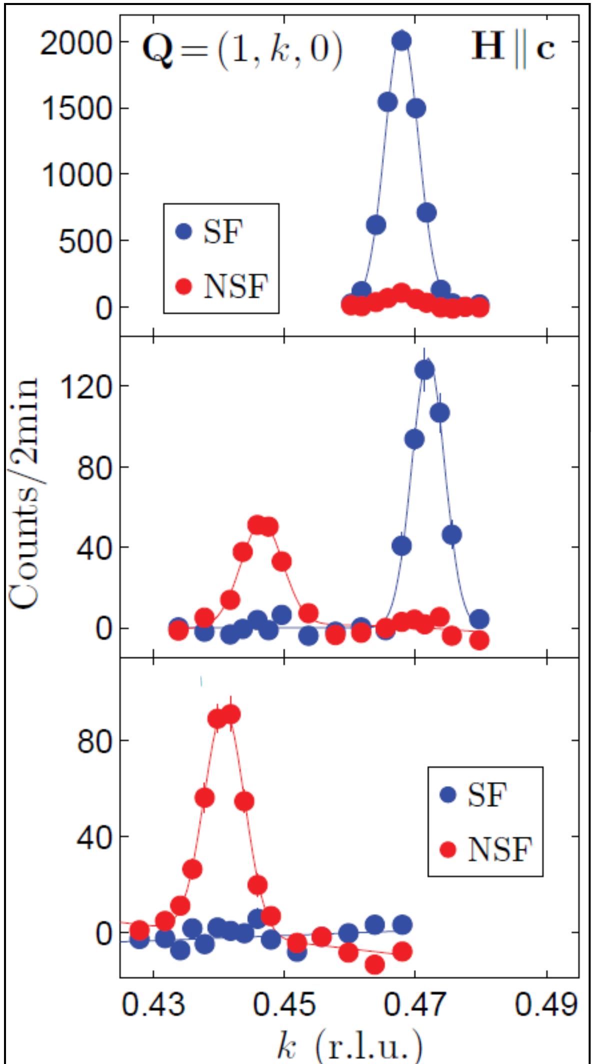
$$\boxed{\tilde{k}_{IC}=[1/2-m_c(H)]/p,\ p=2}$$

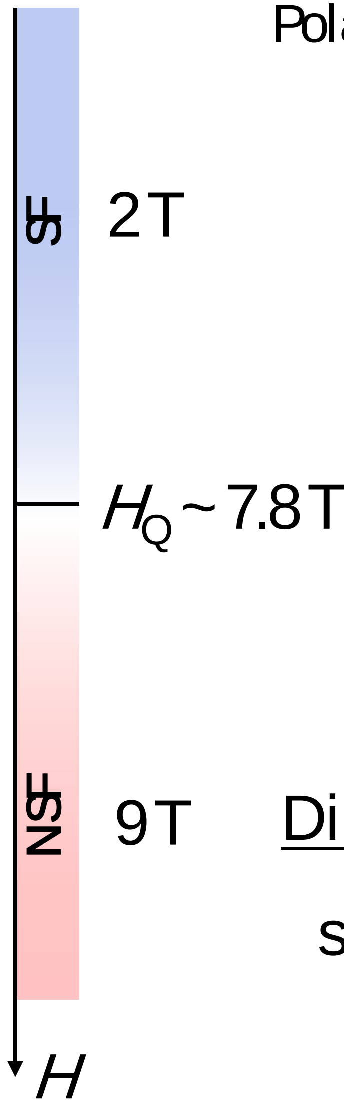
for longitudinal dipolar correlations

Dominant quadrupolar-nematic correlations

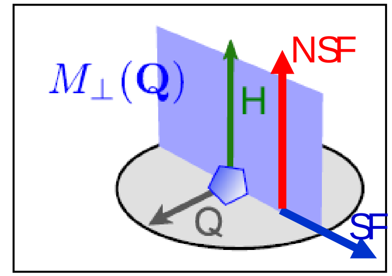
2.2 Elastic neutron scattering above H_Q

❖ 3. Spin components involved in short-range correlations





Polarized neutrons, vertical field, 50 mK



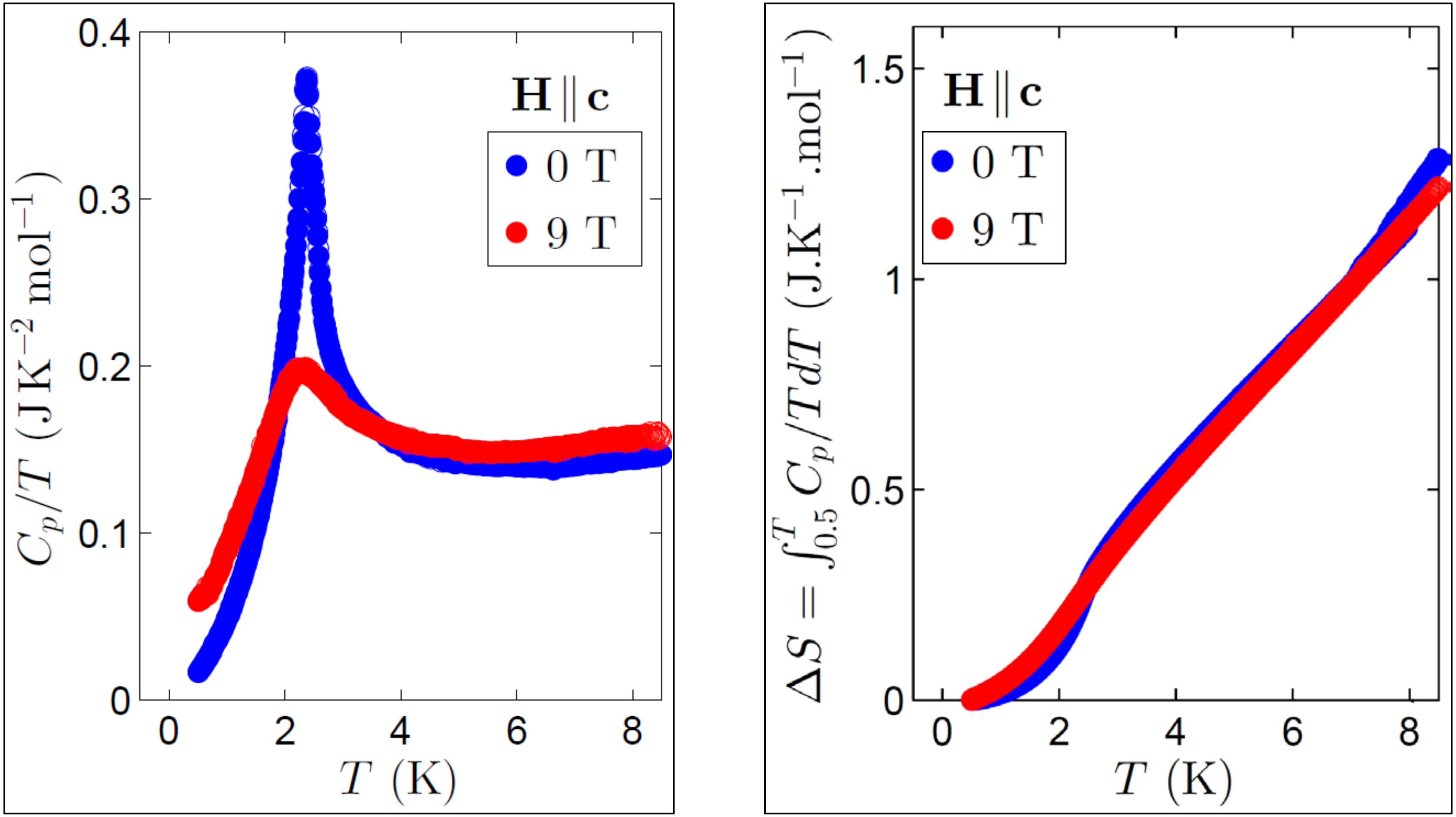
$\boxed{\sigma_{\mathrm{SF}},\ \perp \mathbf{H} \perp \mathbf{Q}}$ In-plane

$\boxed{\sigma_{\mathrm{NSF}},\ \parallel \mathbf{H} \perp \mathbf{Q}}$ Out-of-plane

Dipolar short-range involve only spin components parallel to H

2.2 Elastic neutron scattering above H_Q

❖ 4. Phase-transition evidenced above H_Q



Thermal <u>phase transition</u> of different universality class

2.2 Summary of our findings

❖ **Below H_Q**

1. Dipolar long-range order related to Vector-Chiral order

2. Incommensurate spin components perpendicular to **H**

❖ **Above H_Q ~ 8 T**

1. Short-range dipolar correlations in all directions

2. Driven by quadrupolar-nematic correlations

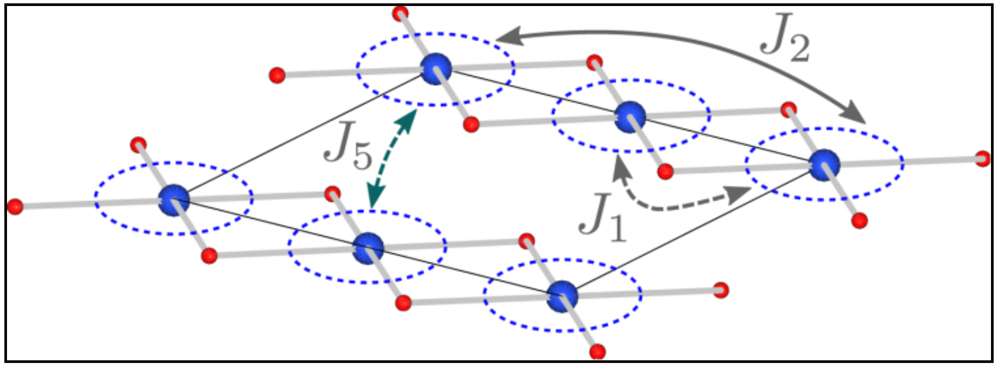
3. Only involve spin components parallel to **H**

4. Thermal phase transition of different universality class

What is the phase above H_Q?

2.3 A Possible Scenario …

❖ Role of frustrated inter-chain interactions



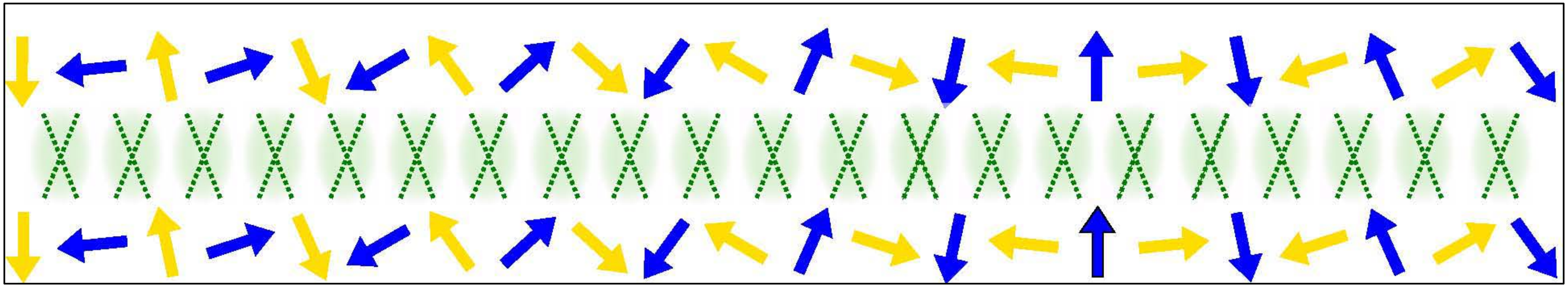
J_1 = -1.6 meV
J_2 = +3.8 meV
J_5 = -0.4 meV

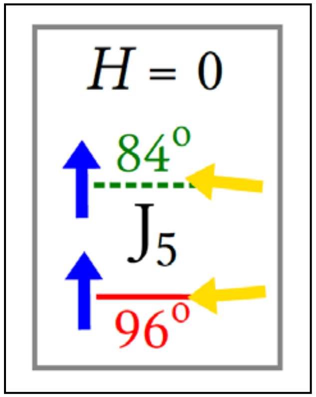
Enderle *et al.*, EPL '05

…Qualitative picture using solitons (fermions) …

2.3 A Possible Scenario …

❖ Role of frustrated inter-chain interactions $H = 0$





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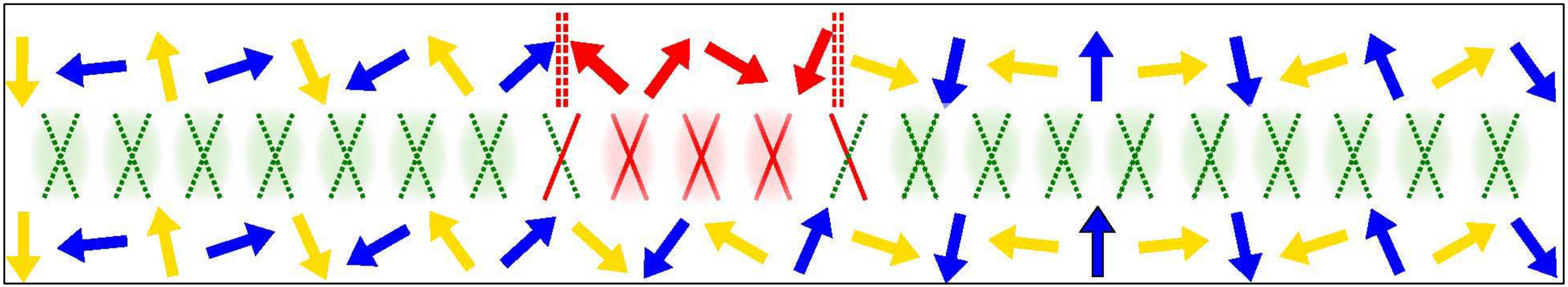
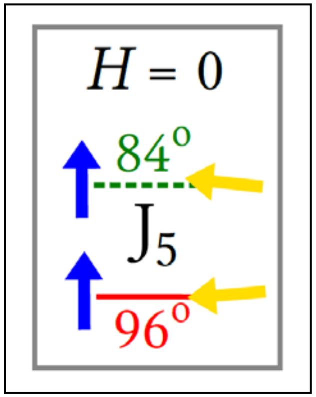


Quantum Fluctuations: 2-soliton + 2-soliton

Furukawa *et al.*, JPSJ'08



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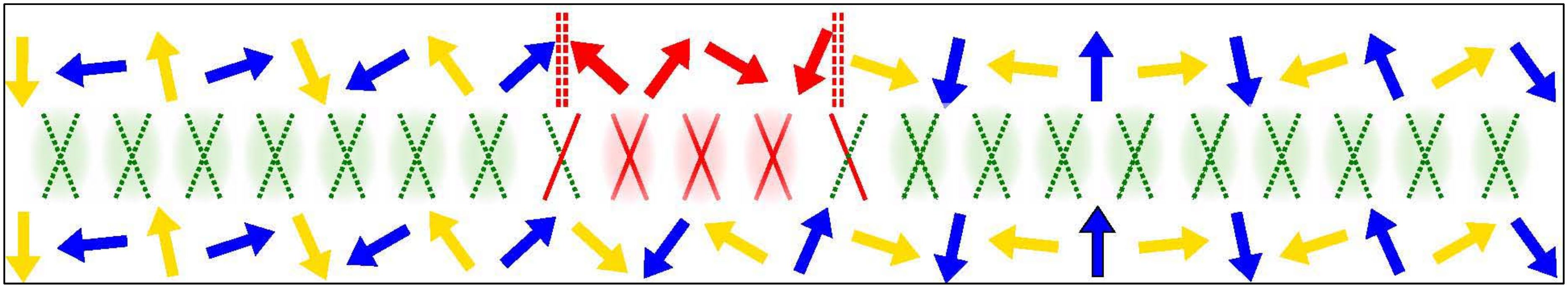
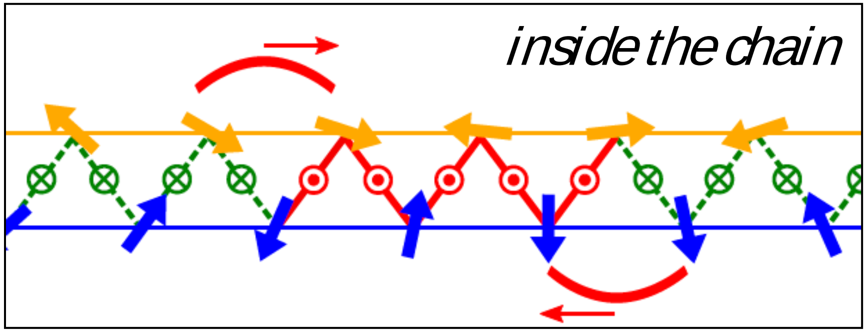
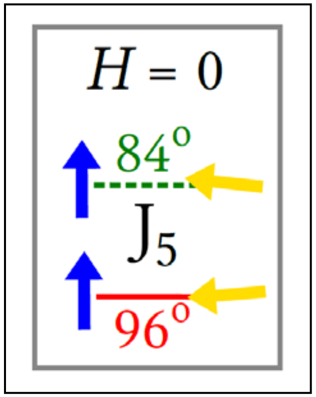
❖ Role of <u>frustrated</u> inter-chain interactions $H = 0$



2-soliton + 2-soliton



A 2-soliton is bound together by FM J_1



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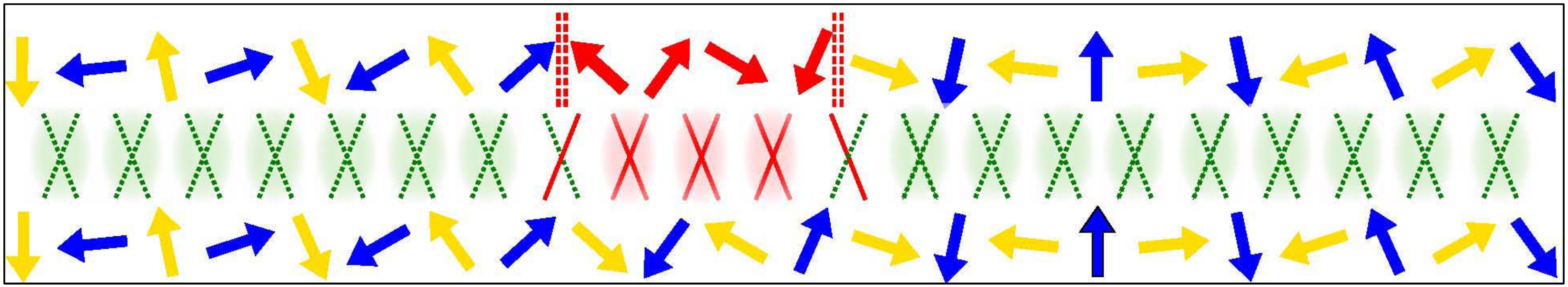

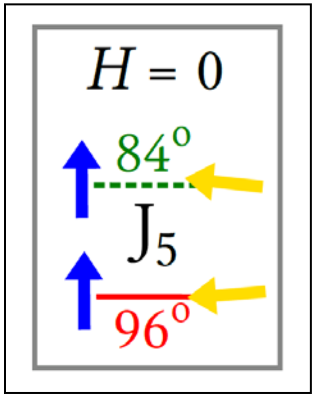

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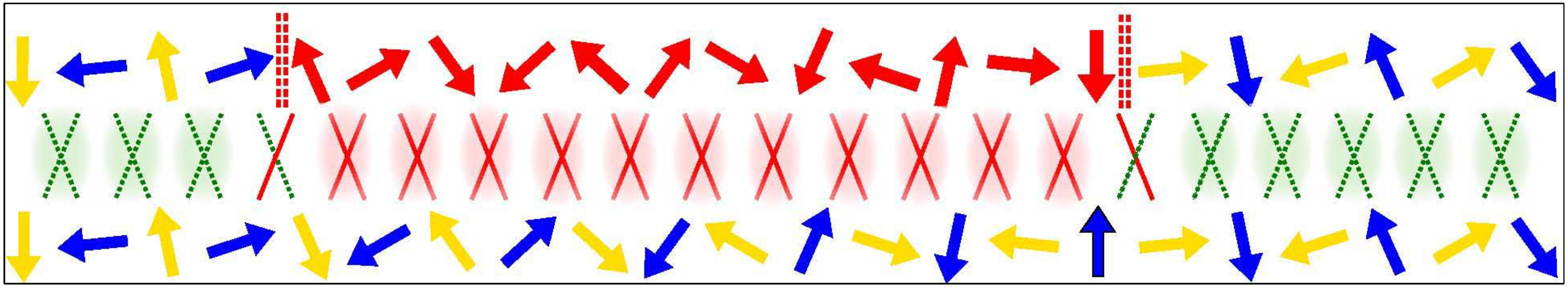
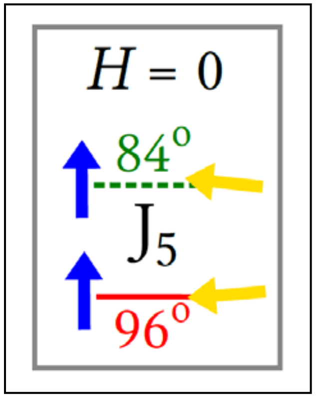
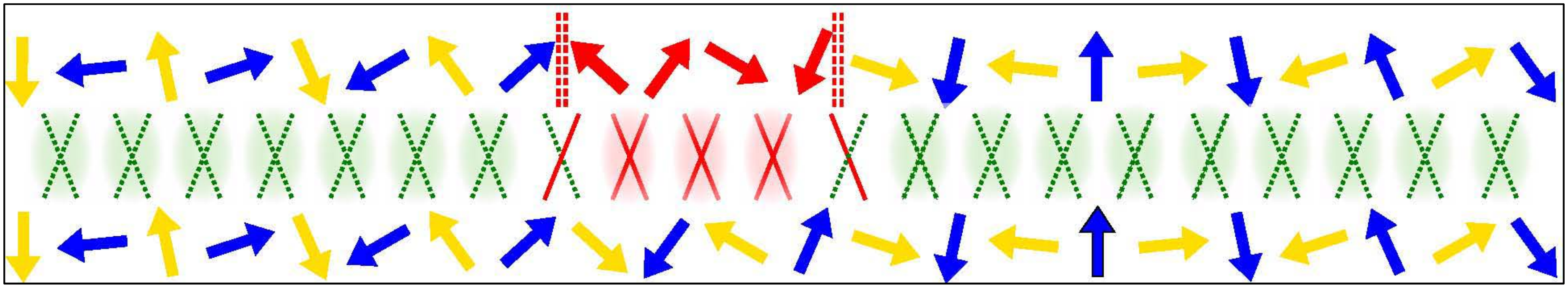


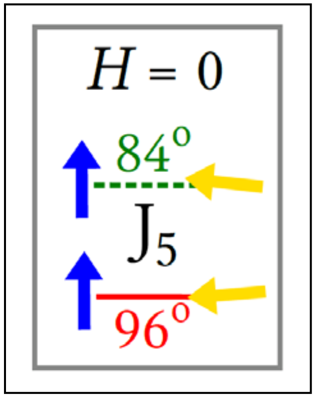


2.3 A Possible Scenario …

❖ Role of <u>frustrated</u> inter-chain interactions $H = 0$



Bound 4-soliton



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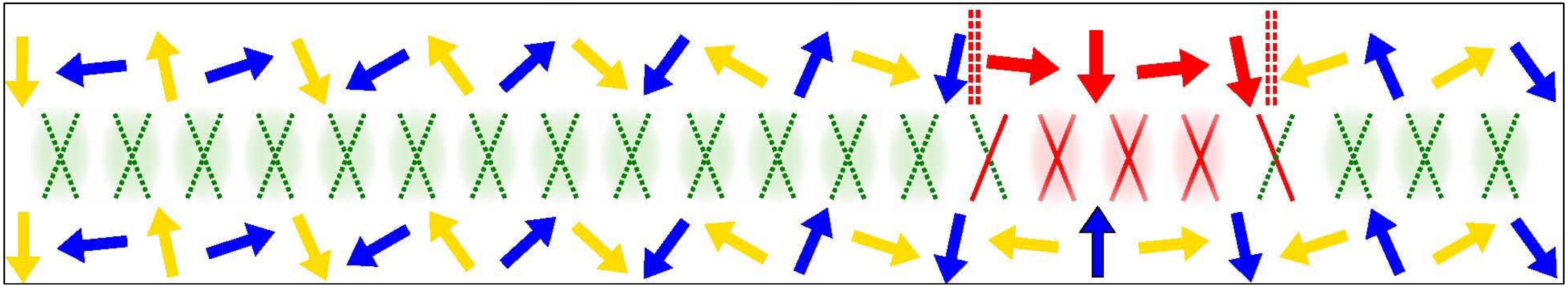

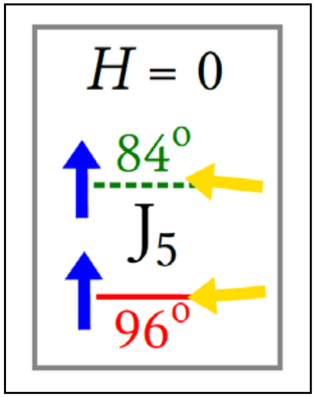

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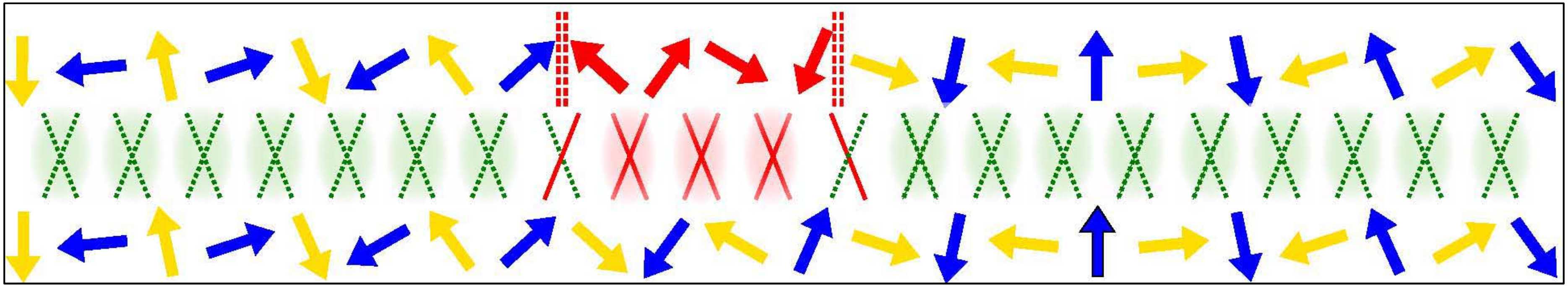


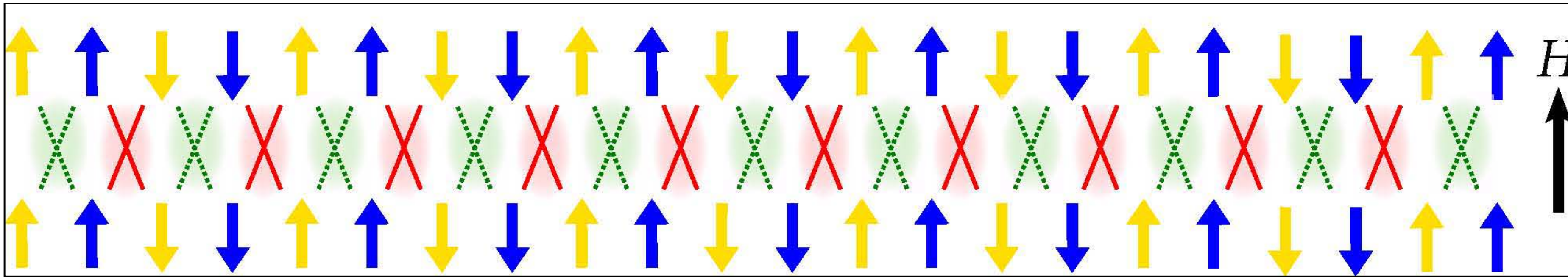
In H=0, long-range dipolar and vector-chiral orders are preserved

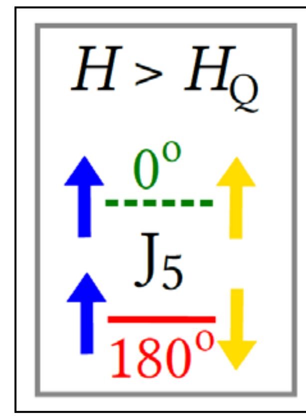
2-soliton bound by intra-chain J_1 → vector-chiral order

4-soliton bound by inter-chain J_5 → long-range dipolar order

2.3 A Possible Scenario …

❖ Role of frustrated inter-chain interactions $H > H_Q$





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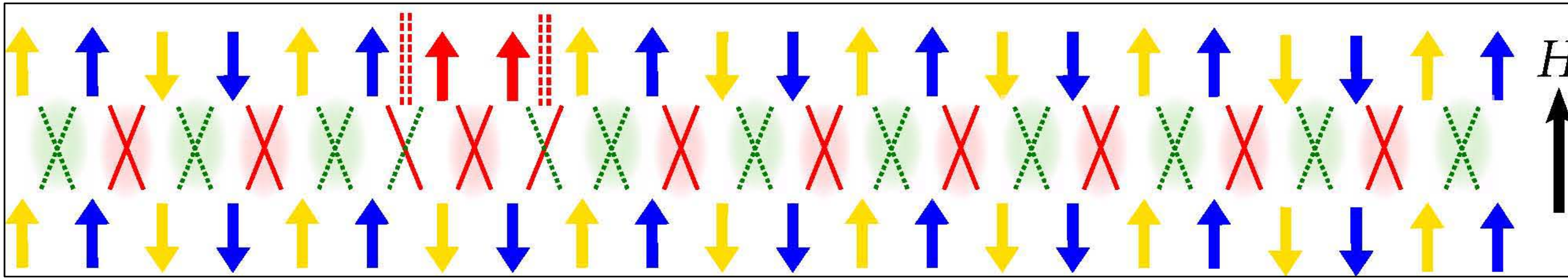
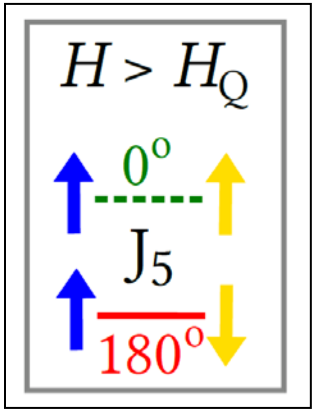


2-soliton + 2-soliton



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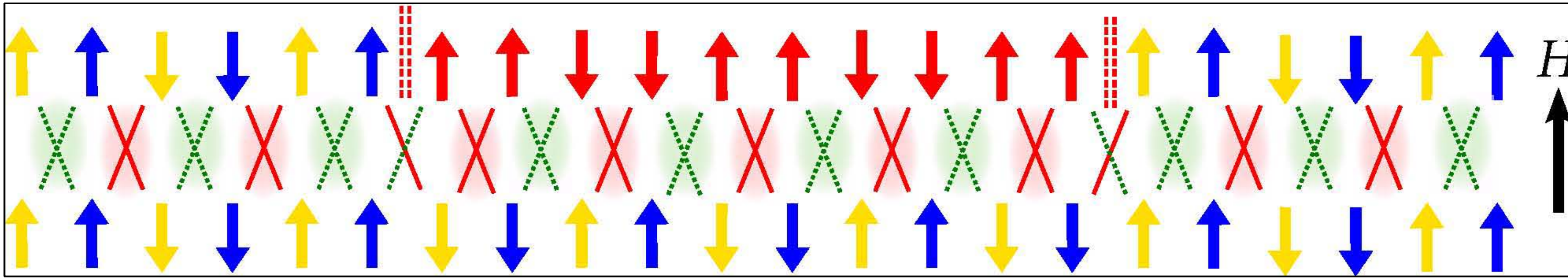
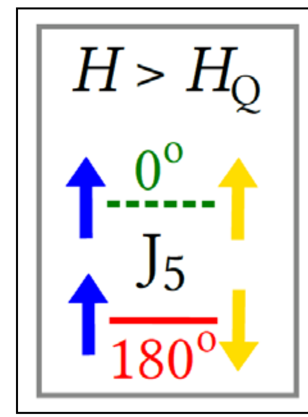
❖ Role of frustrated inter-chain interactions $H > H_Q$





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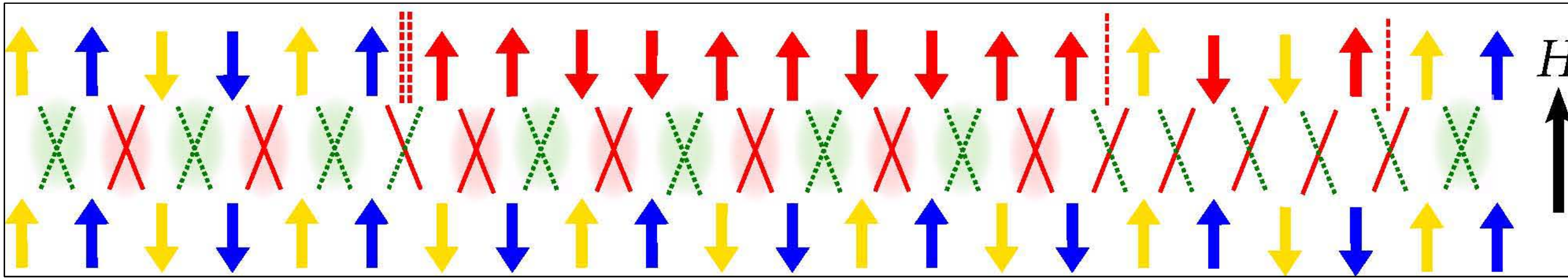

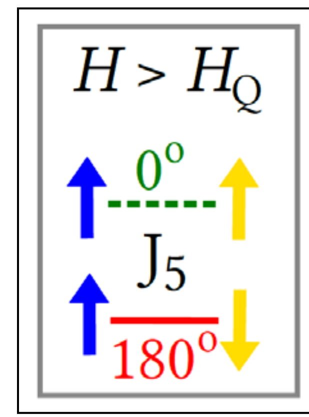

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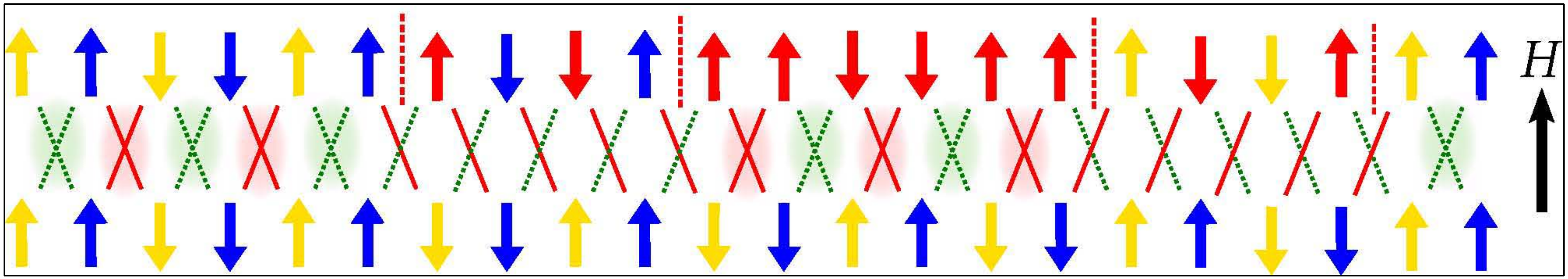

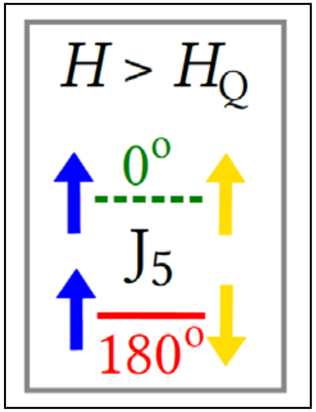

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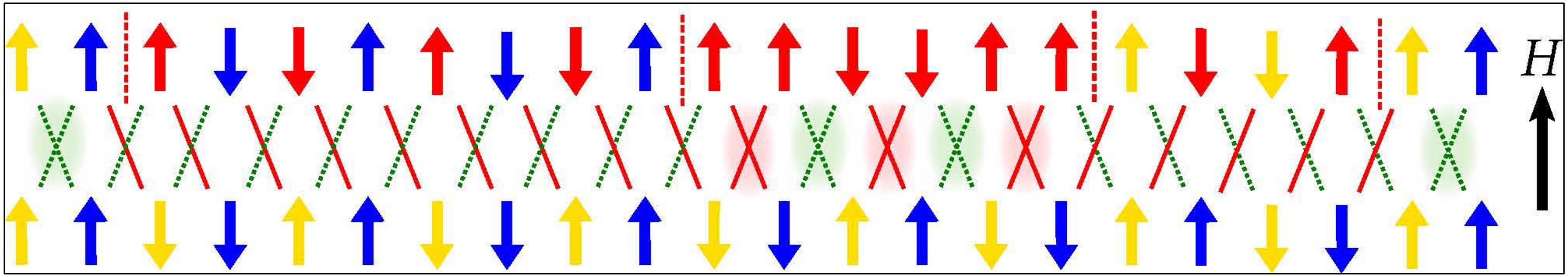
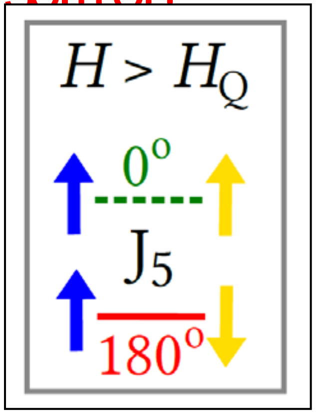


1-soliton 1-soliton 1-soliton 1-soliton



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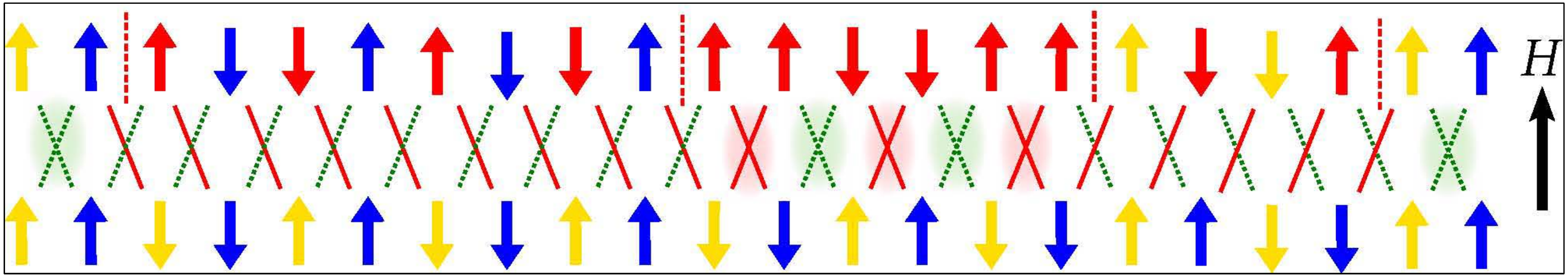
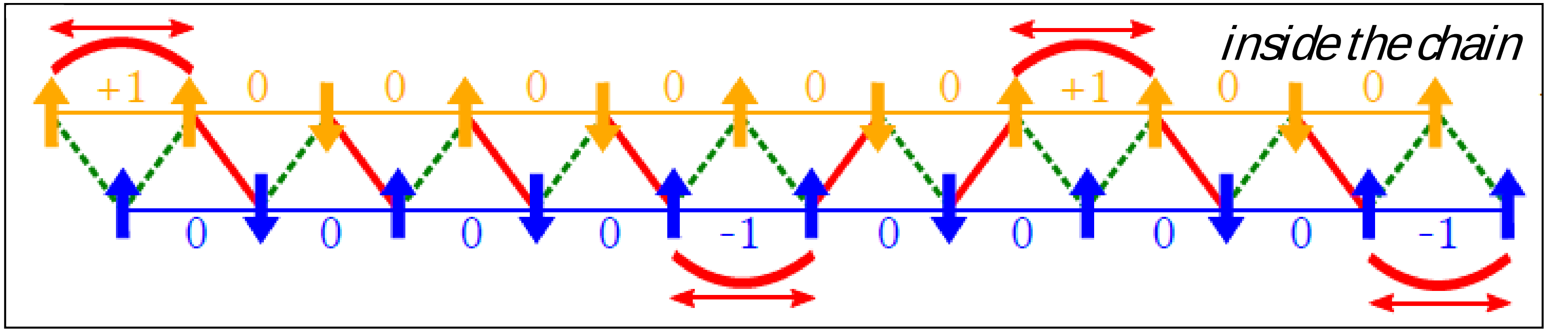
❖ Role of frustrated inter-chain interactions $H > H_Q$



In $H>H_Q$ long-range dipolar order is destroyed

However, there is a non-local positional order (“nematic”)



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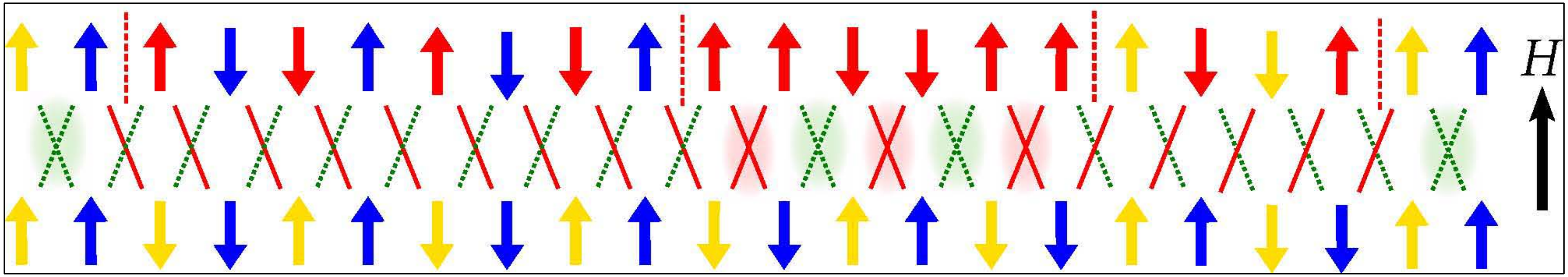


In $H > H_Q$ long-range dipolar order is destroyed

However, there is a non-local positional order ("nematic")

Conclusions

1. A "spin-liquid" is stabilized above H_Q by frustrated inter-chain inter.

2. Different from dipolar LR below H_Q and quadrupolar LR above H_C

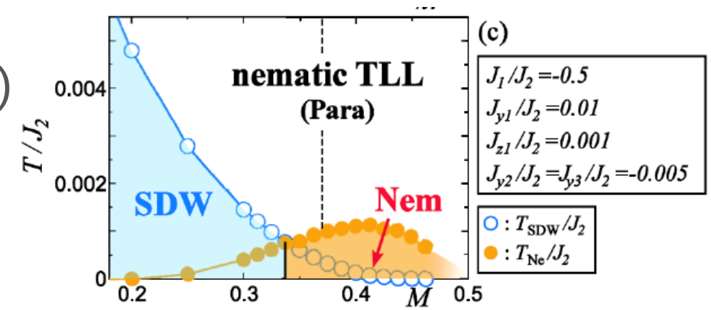
Outlook

❖ $LiCuVO_4$

Nature of the phases above H_Q and H_C remains to be clarified

High density of two-magnon pairs in presence of frustrated interactions

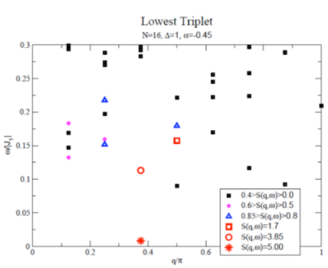
Bosonization+DMRG (Sato, Hikihara, Momoi ArXiV'12)



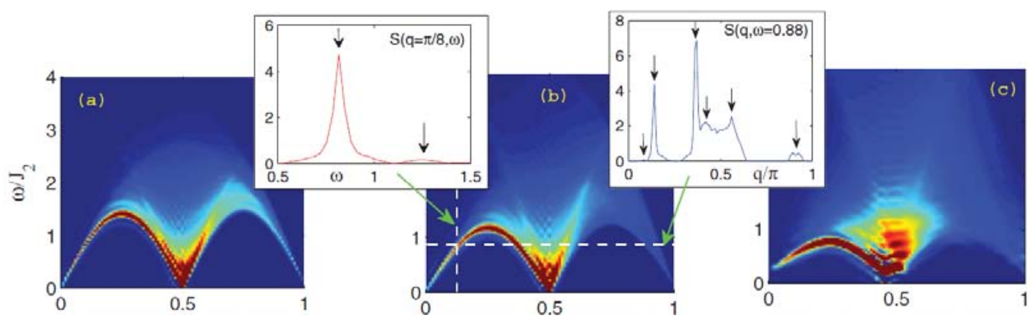
❖ $LiCuSbO_4$

How to model such a complex powder spectrum in pure 1D system?

Exact Diagonalization (Kumar, Soos)



Time-Dependent DMRG ? (Ren and Sirker, PRB'12)



Thank you for your attention

Lascaux Cave (17000 BC), Montignac-sur-Vézère, South-West France