INSTITUTE FOR QUANTUM MATTER

A collaboration between
JOHNS HOPKINS UNIVERSITY
and PRINCETON UNIVERSITY

Neutron Scattering from Quantum and Frustrated Spin Chains

Martin Mourigal

Institute for Quantum Matter

Johns Hopkins University, Baltimore, USA

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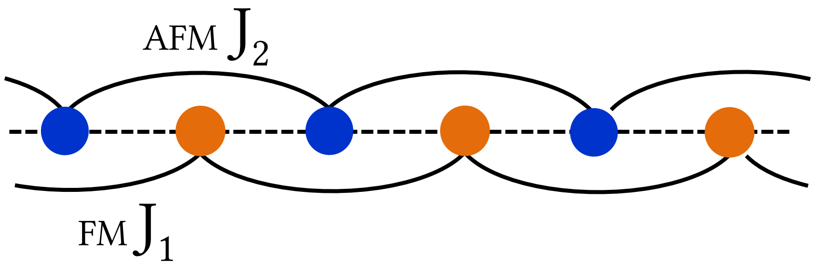
JOHNS HOPKINS UNIVERSITY

Outline

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

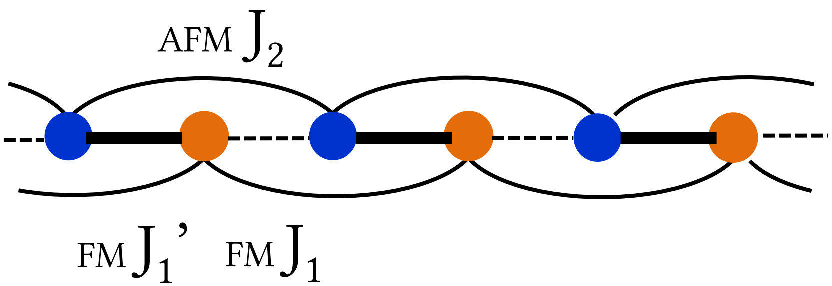
1. Motivation

2. Frustrated ferromagnetic chains in $LiCuVO_4$



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

3. Frustrated chains in powder-sample of $LiCuSbO_4$



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

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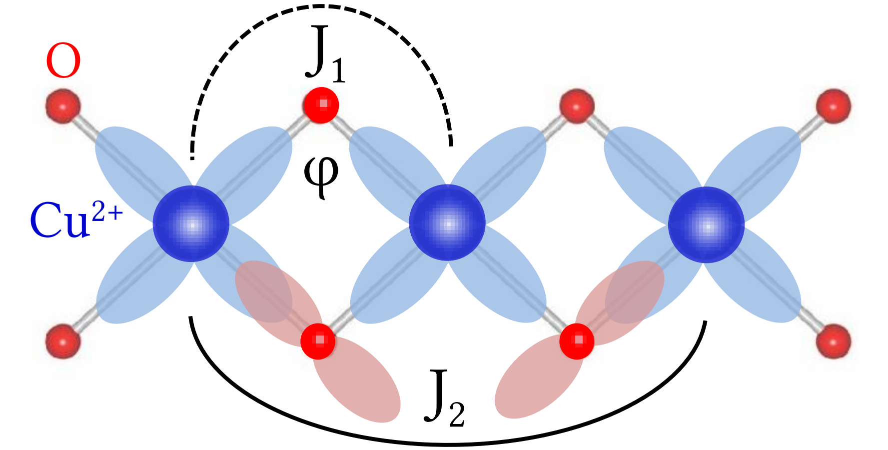
Framework: (Quasi-) 1D Heisenberg quantum ($S = 1/2$) spin chains

1. Motivation

1.1 Edge-sharing cuprate chains

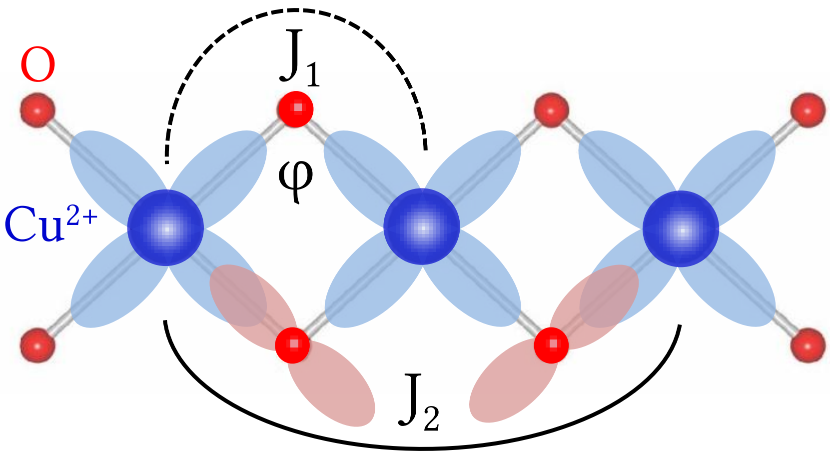


Jahn-Teller distorted CuO_6 octahedral

J_1 can be F or AF ($\varphi \sim 90^\circ$), J_2 is AF

$LiCu_2O_2$, $LiCuSbO_4$, $Rb_2Cu_2Mo_3O_{12}$, ...

1.1 Edge-sharing cuprate chains



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J_1 can be F or AF ($\varphi \sim 90°$), J_2 is AF

$LiCu_2O_2$, $LiCuSbO_4$, $Rb_2Cu_2Mo_3O_{12}$, …

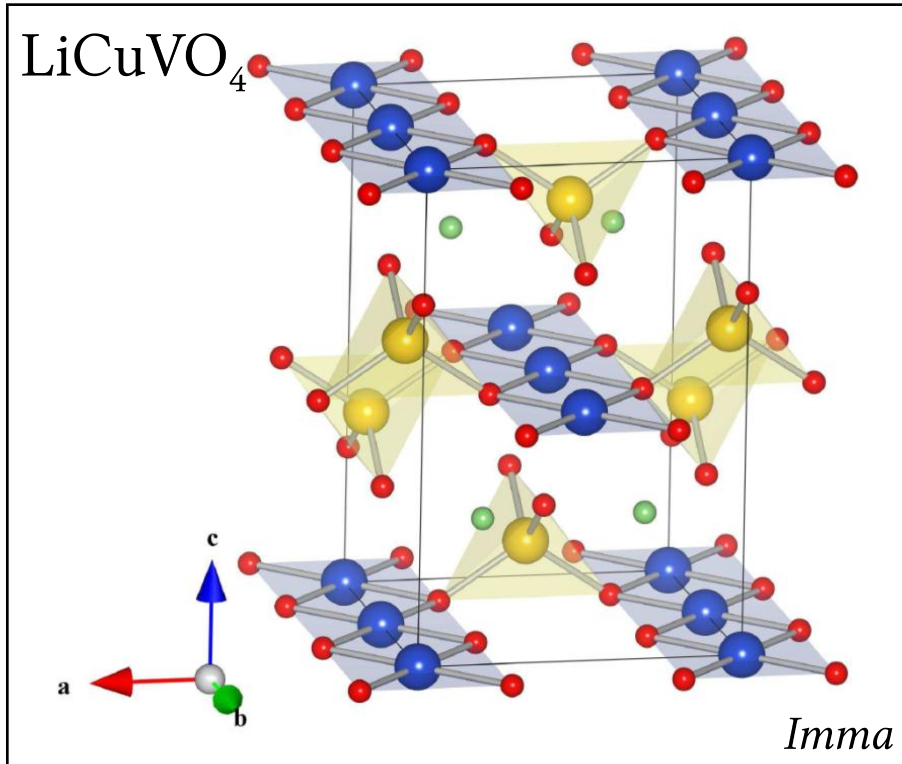
Chains along *b*, symmetric struc.

J_1 =1.6 meV (F), J_2 =5.60 meV (AF)

Weak interchain interactions

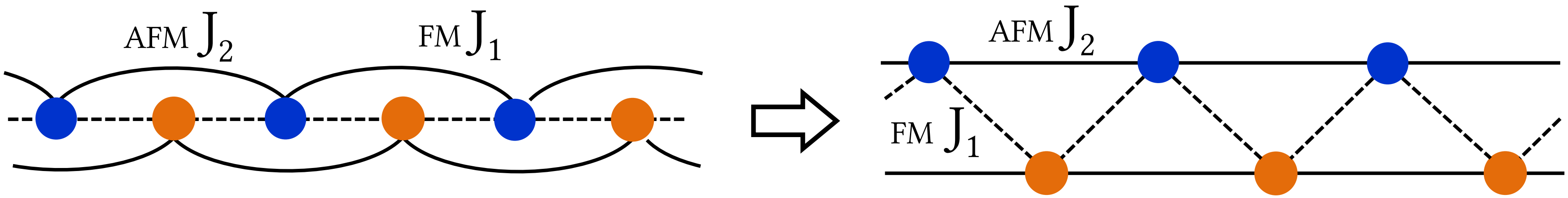
J_5 =0.40 meV (AF)

Néel ordering at T_N = 2.4K



See: La Fontaine *et al.*, Acta Cryst. '89 | H.-J. Koo *et al.* Inorg. Chem. '11 | Enderle *et al.* EPL '05

1.2 The spin-1/2 frustrated chain model



Easy-plane anisotropy Δ

U(1) symmetry

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

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

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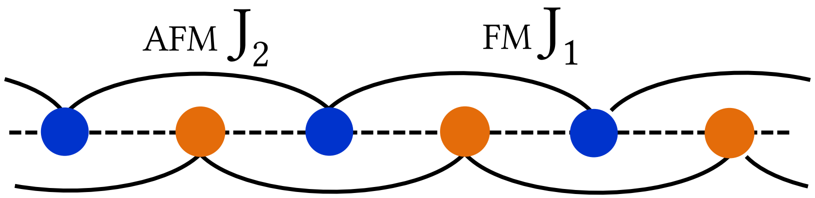

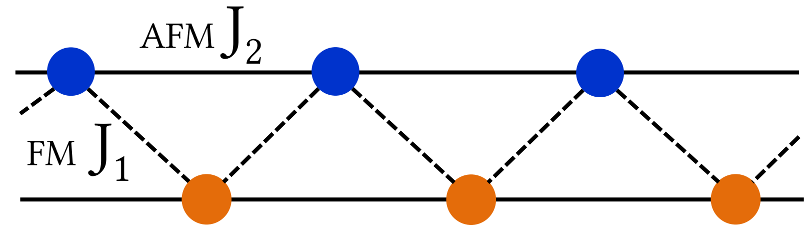

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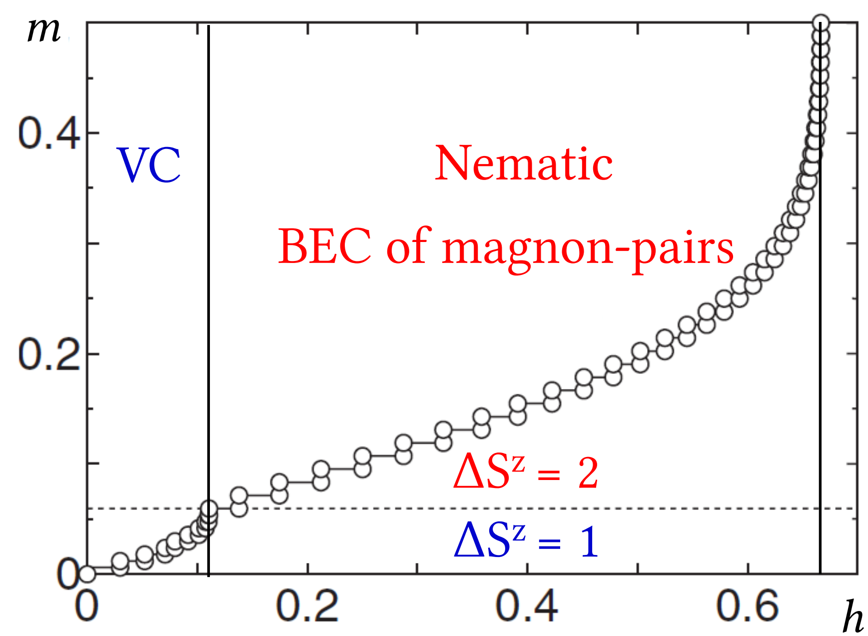
DMRG for pure 1D with J_2/J_1=-0.5



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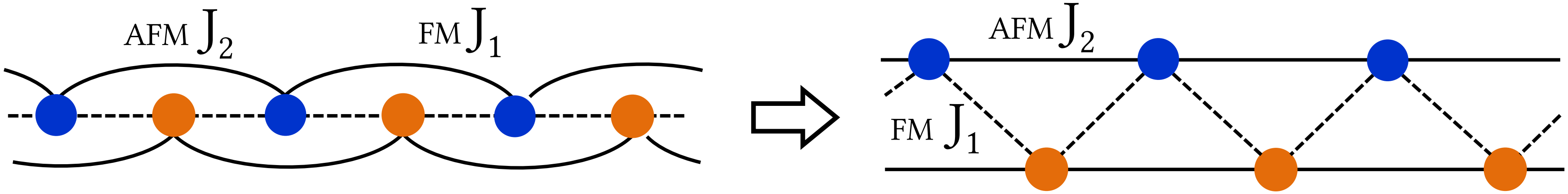

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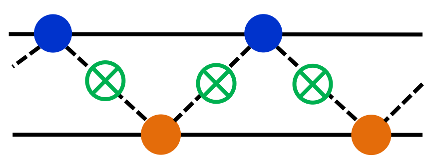
Vector-Chiral (VC)

$\langle \sin(\theta_i - \theta_{i+1}) \rangle \neq 0$

relative-angle



$\langle S_0^+ S_i^- \rangle$	algebraic, incomm.
$\langle S_0^z S_i^z \rangle$	algebraic
$\langle S_0^+ S_1^+ S_i^- S_{i+1}^- \rangle$	exponential

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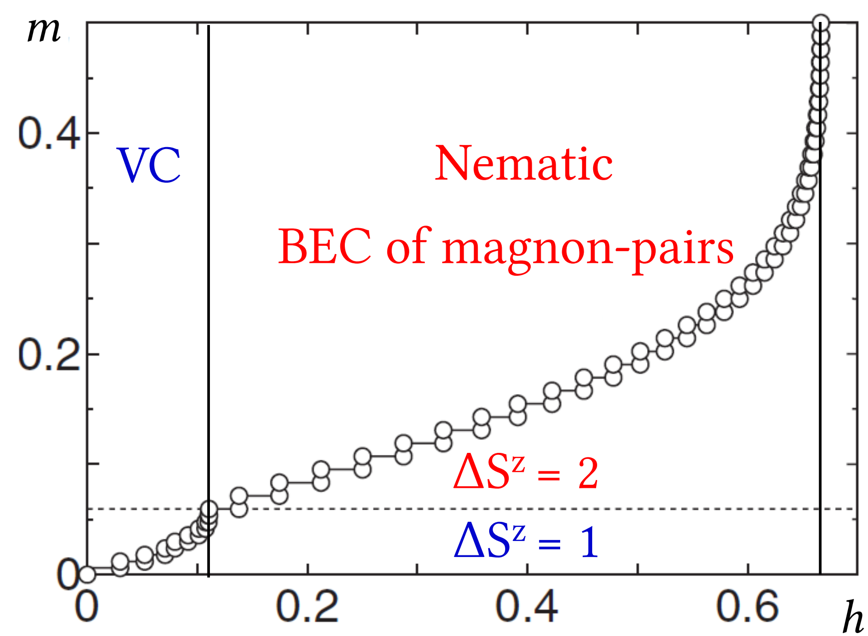

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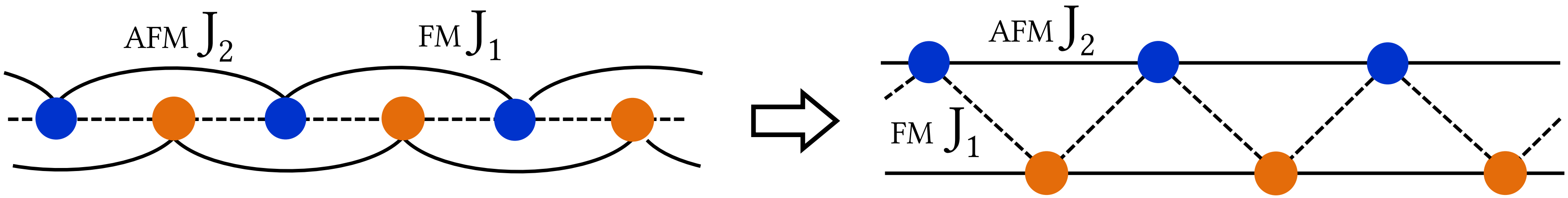

Easy-plane anisotropy Δ

U(1) symmetry

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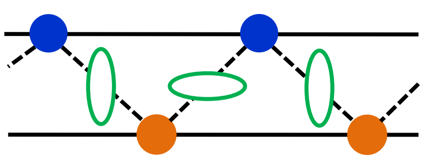
Quadrupolar Nematic (Ne)

$\langle \cos(\theta_i + \theta_{i+1}) \rangle \neq 0$

center-of-mass



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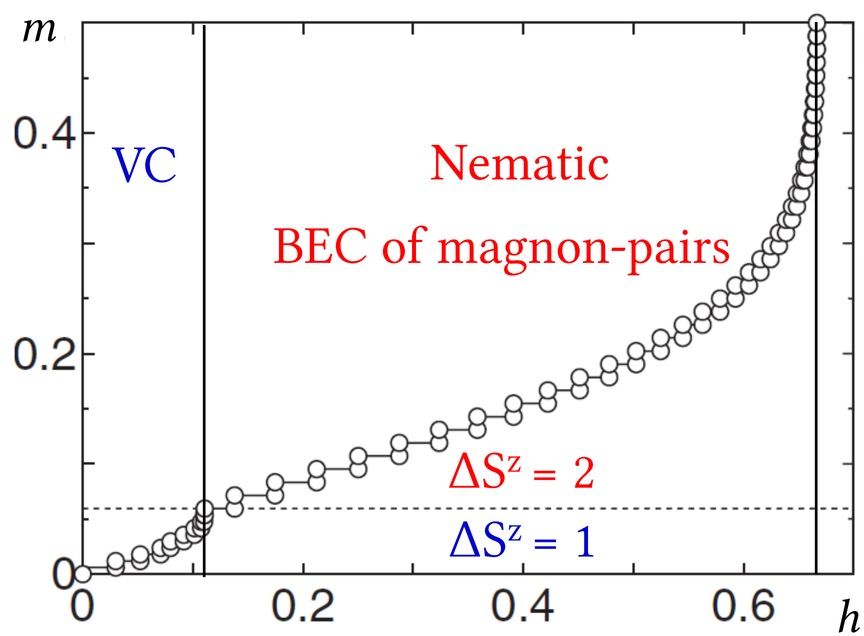

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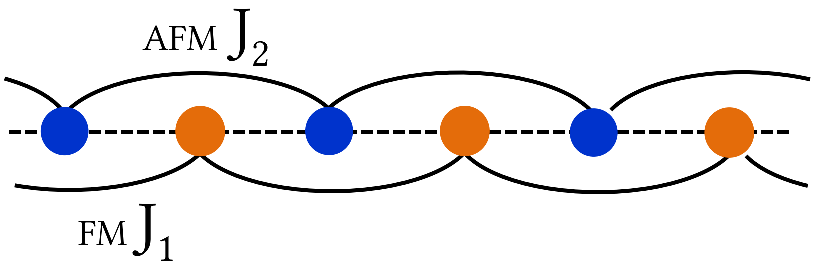

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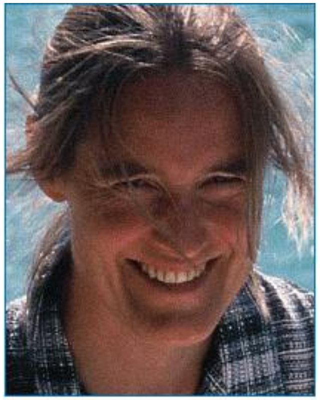
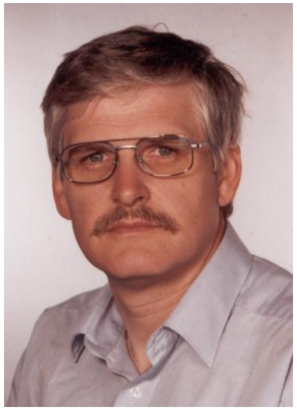


M. Enderle
(ILL, Grenoble)

B. Fåk
(CEA, Grenoble)



R. Kremer
(Max-Planck, Stuttgart)

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

A. Prokoviev (Vienna)

A. Schneidewind (Munich)

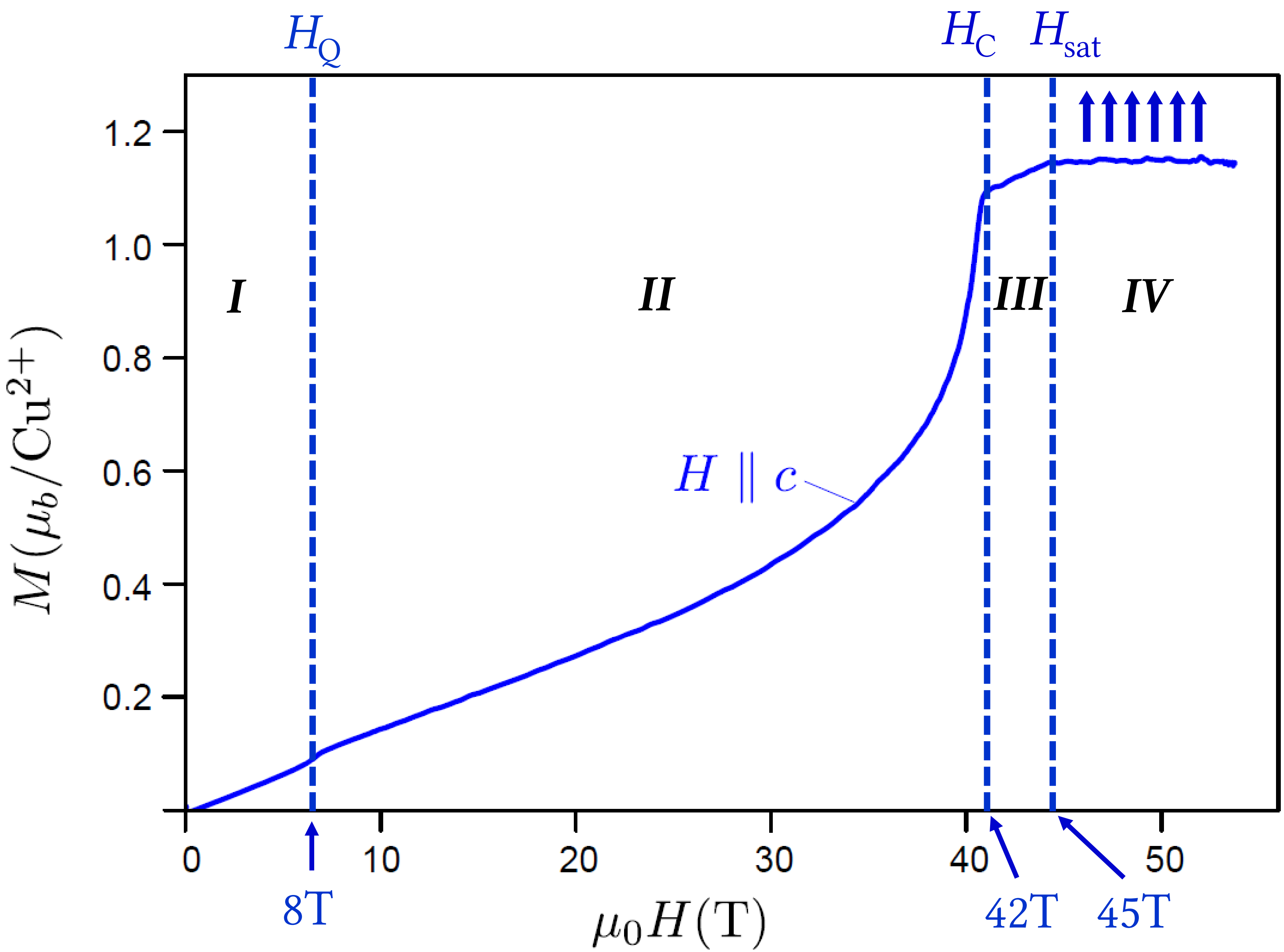
A. Hiess (ESS, Lund)

2.0 Magnetization curve of $LiCuVO_4$

Can quadrupolar-nematic phases be observed ?

What is the effect of quasi-1Dness ?

2.0 Magnetization curve of $LiCuVO_4$



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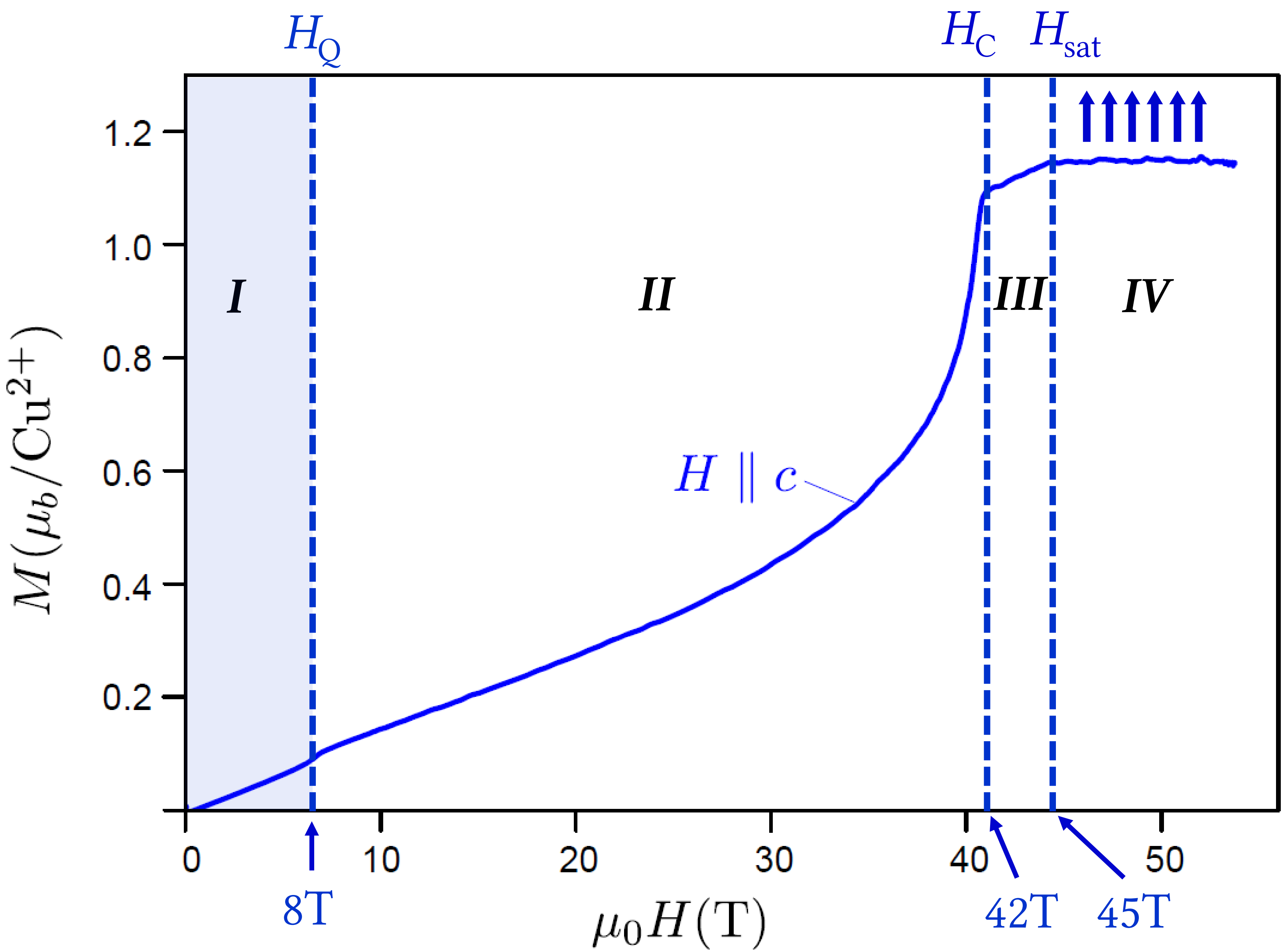

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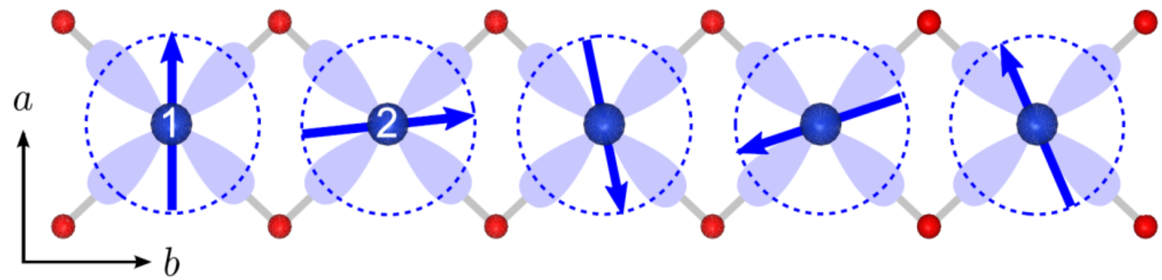
2.1 Below H_Q=8T

❖ Dipolar long-range order

Model spin-cycloid T_N =2.4 K

Ferroelectric behavior



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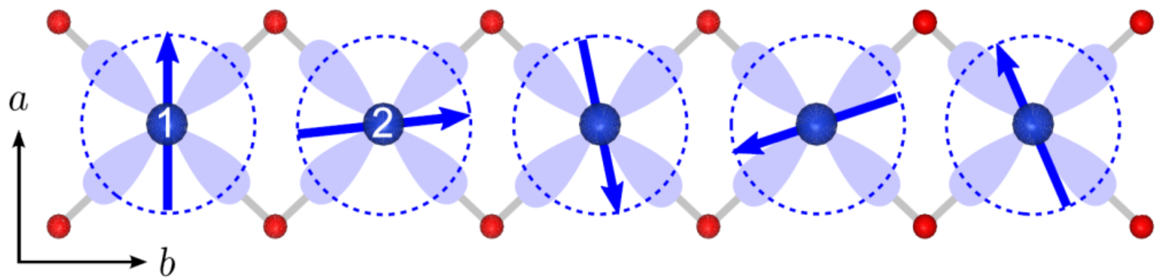

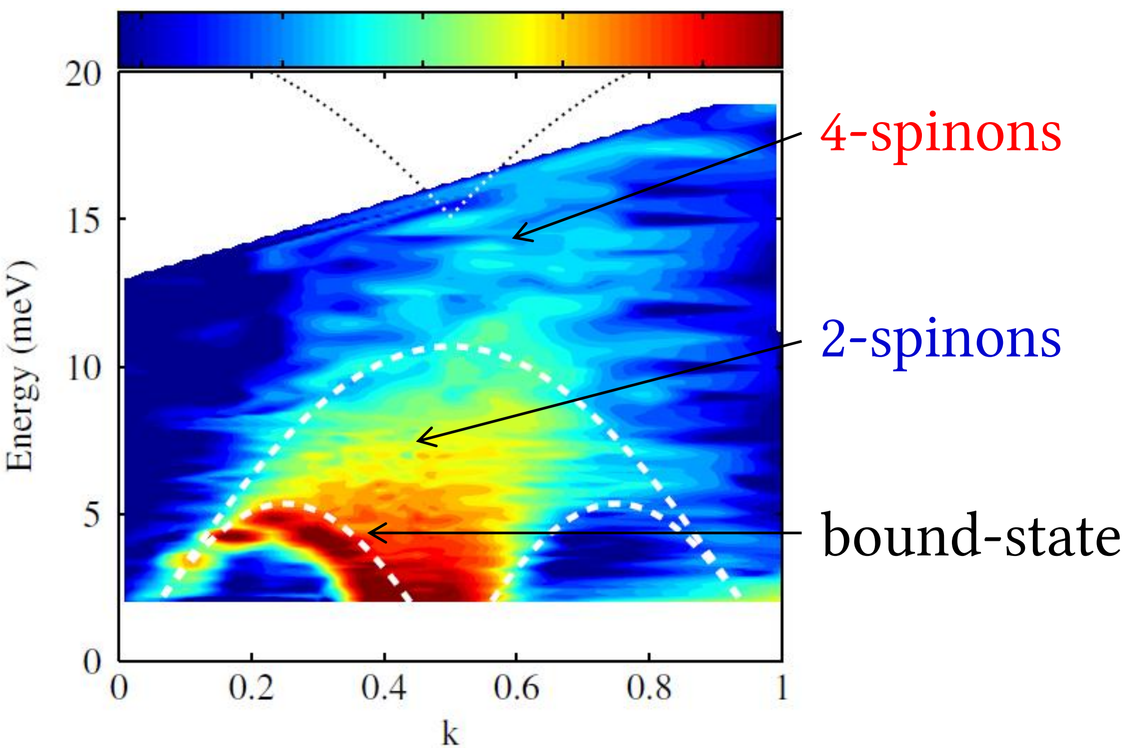

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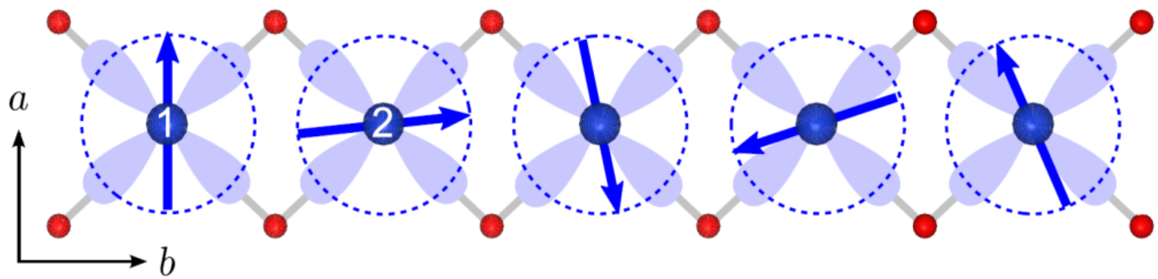

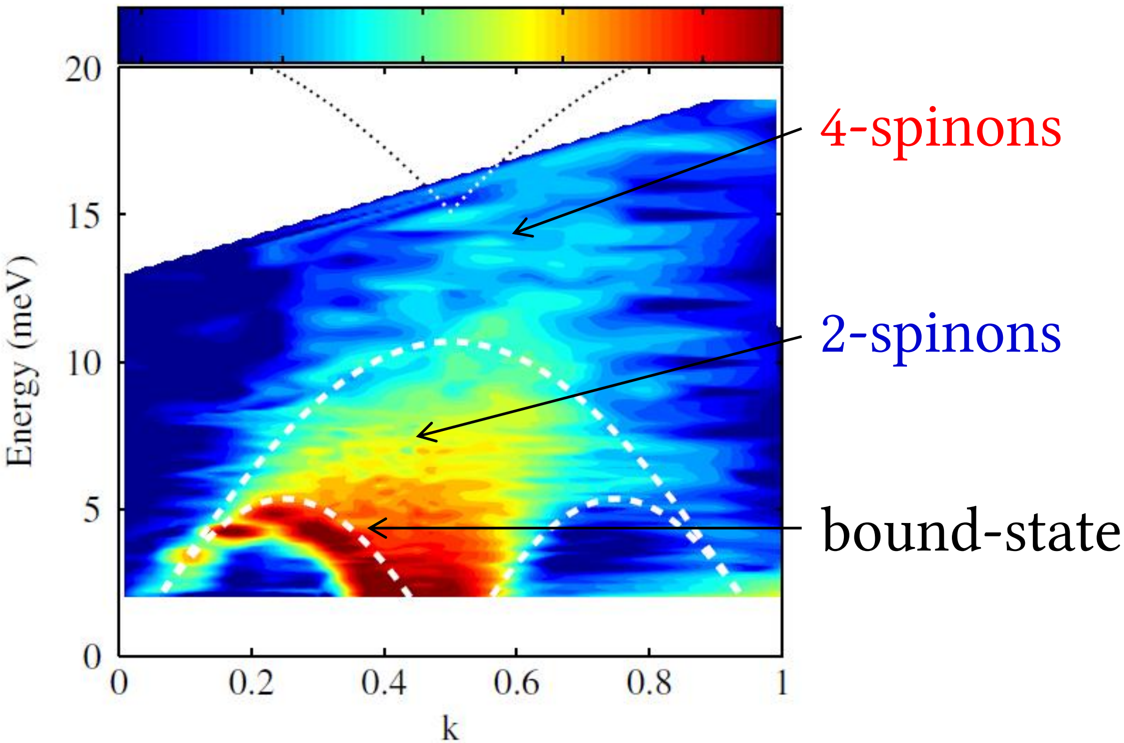

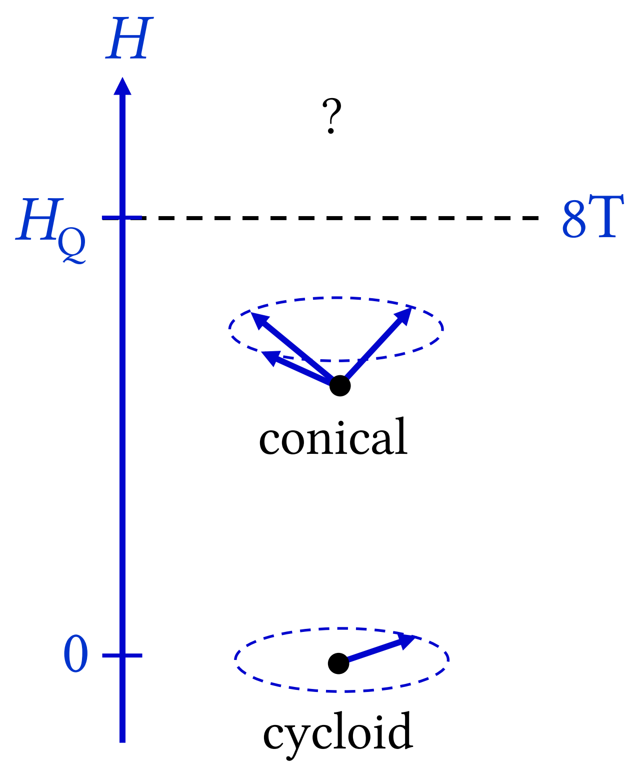

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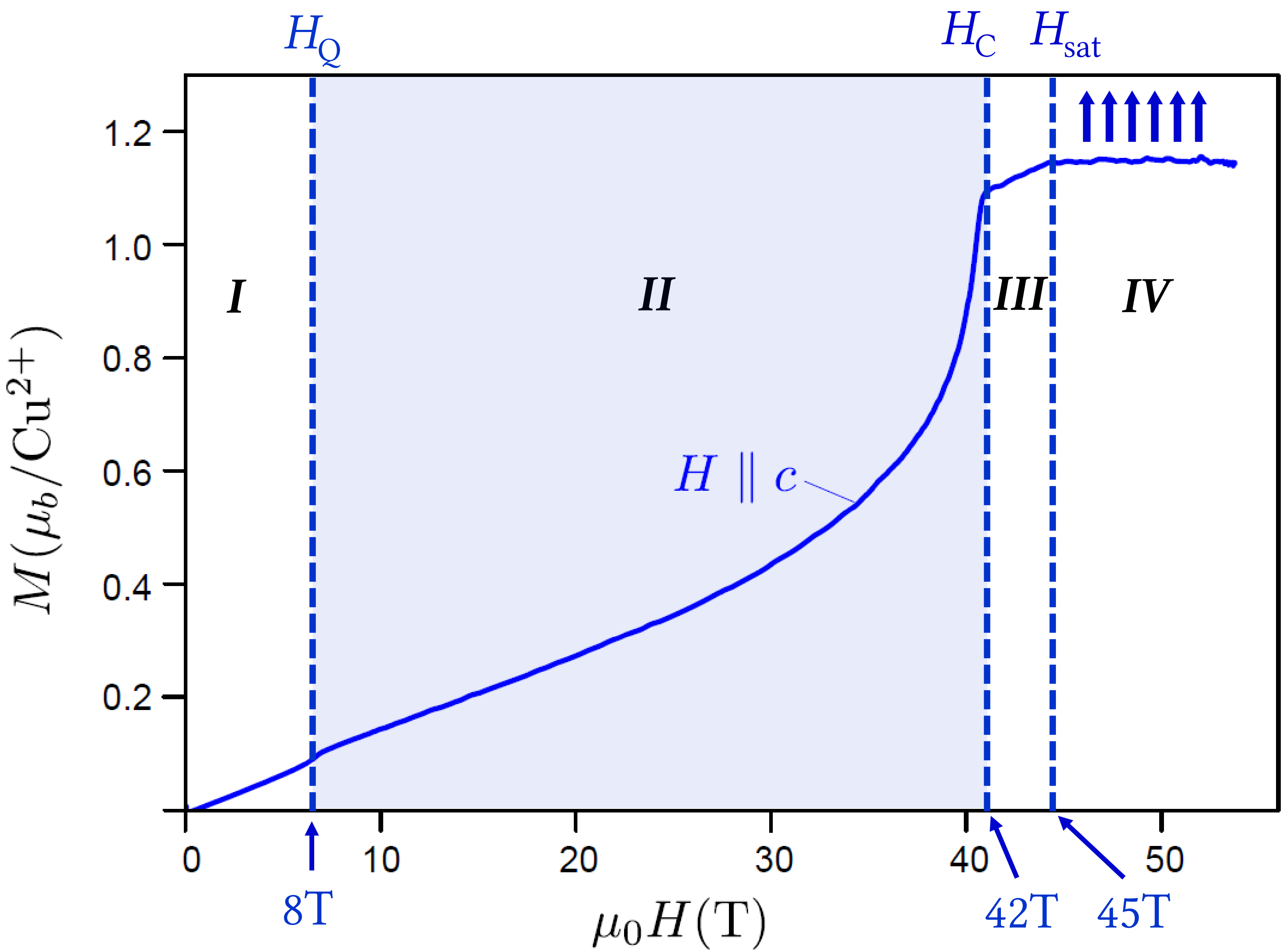

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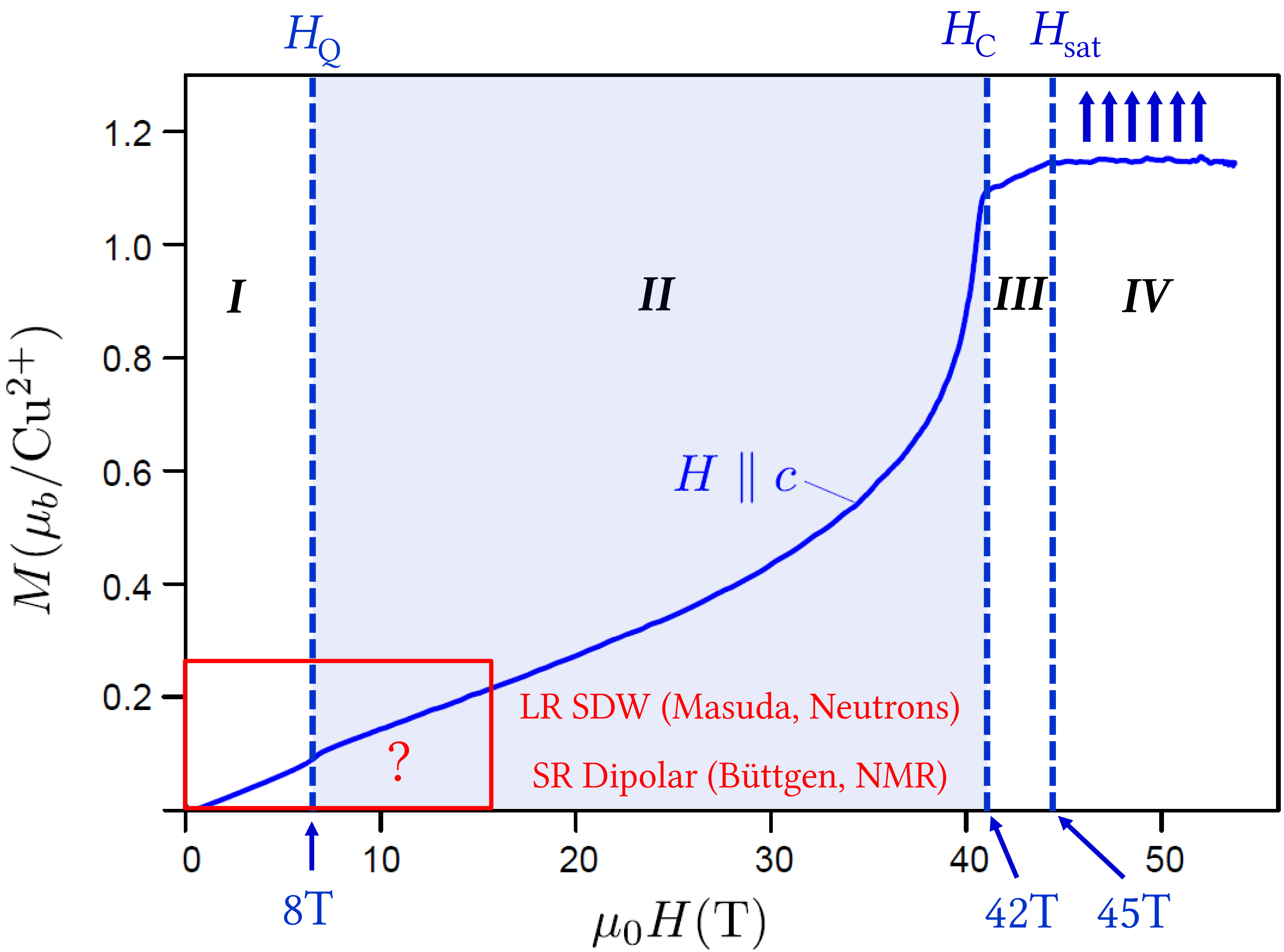

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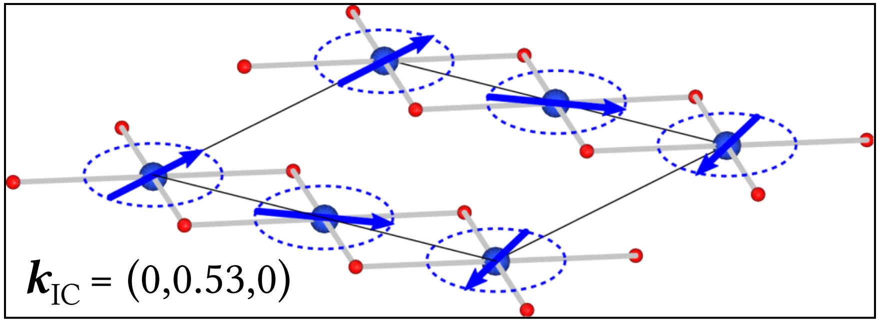
❖ 1. Dipolar spin correlations become short-ranged



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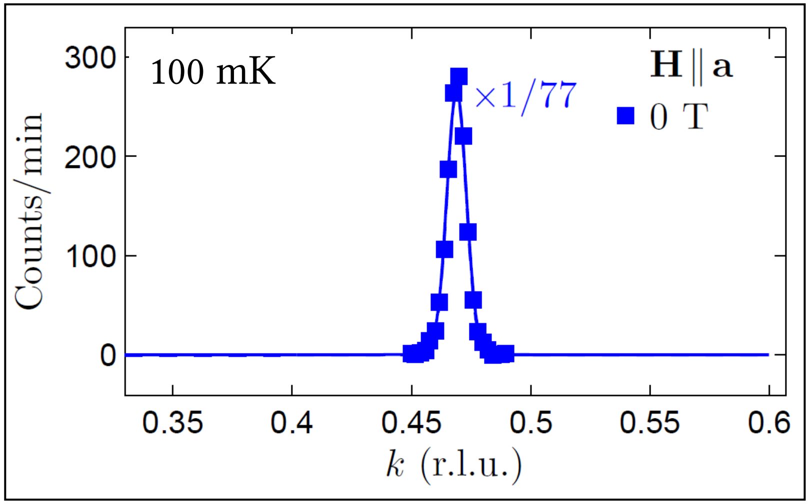
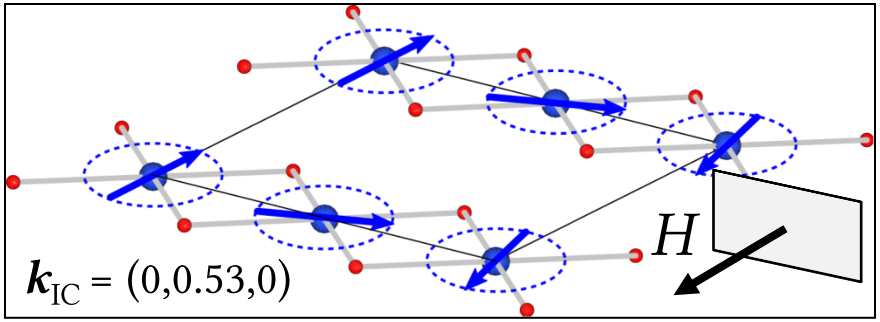


bc-plane, $\boldsymbol{Q}$ = (0,1,1) - $\boldsymbol{k}_{IC}$



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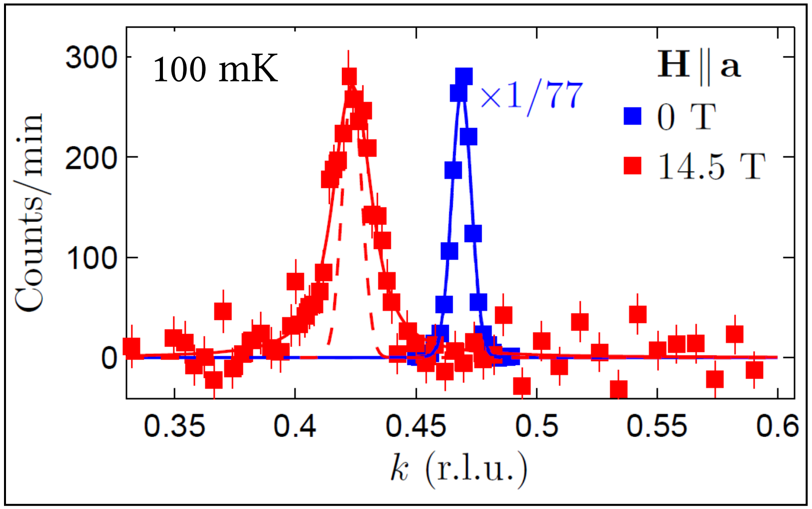

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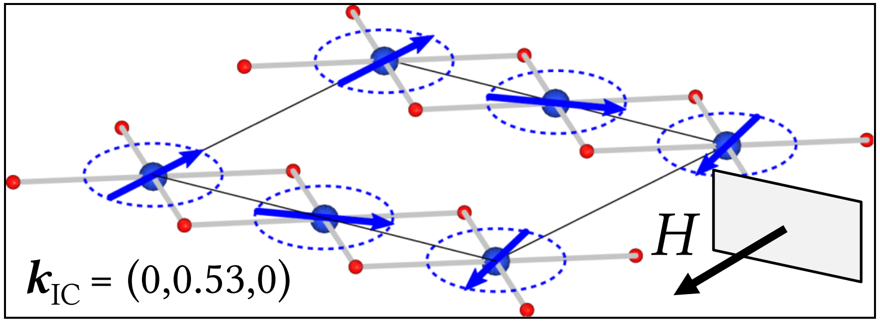

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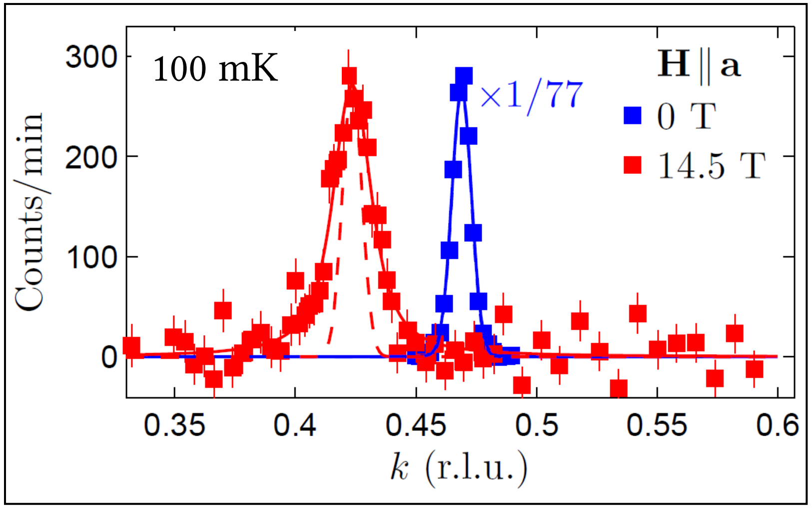

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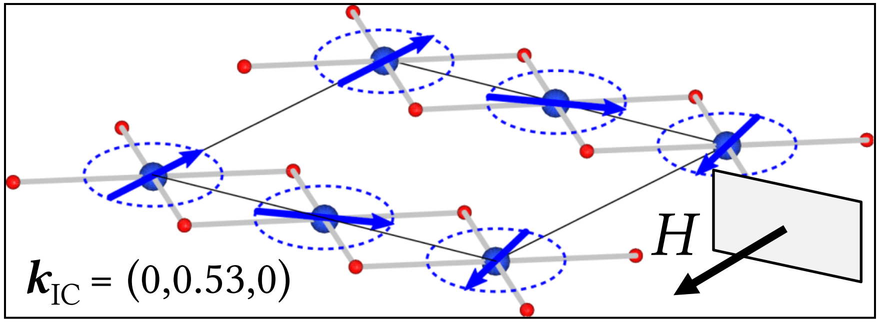

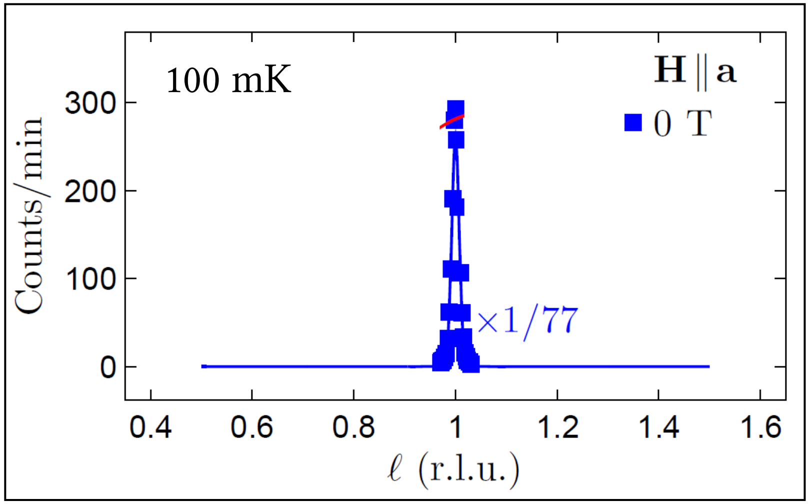

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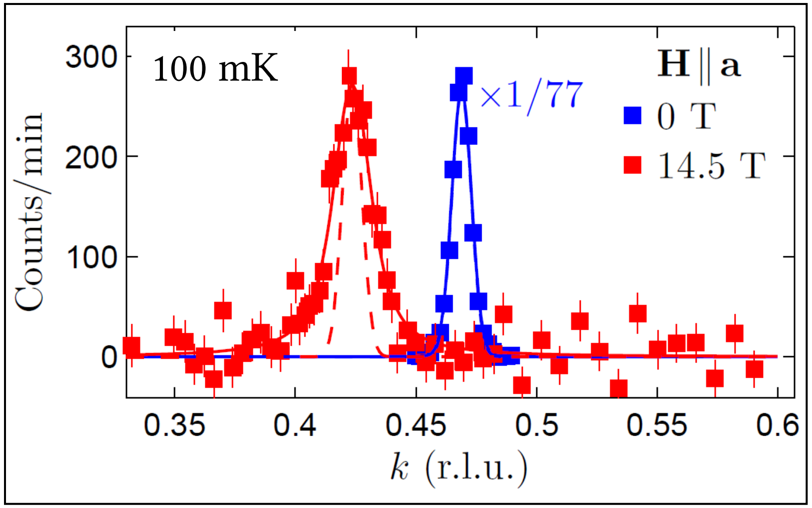

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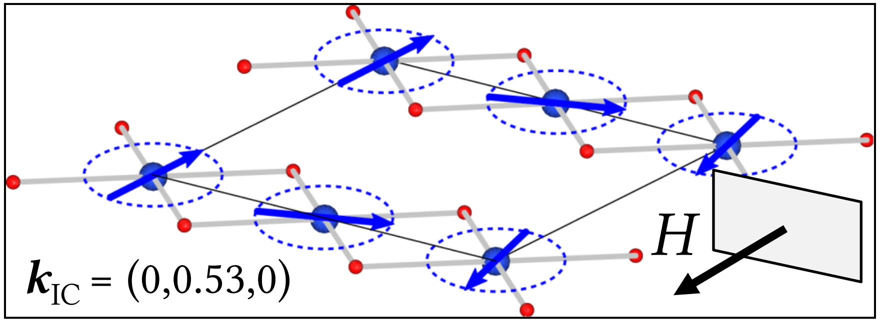

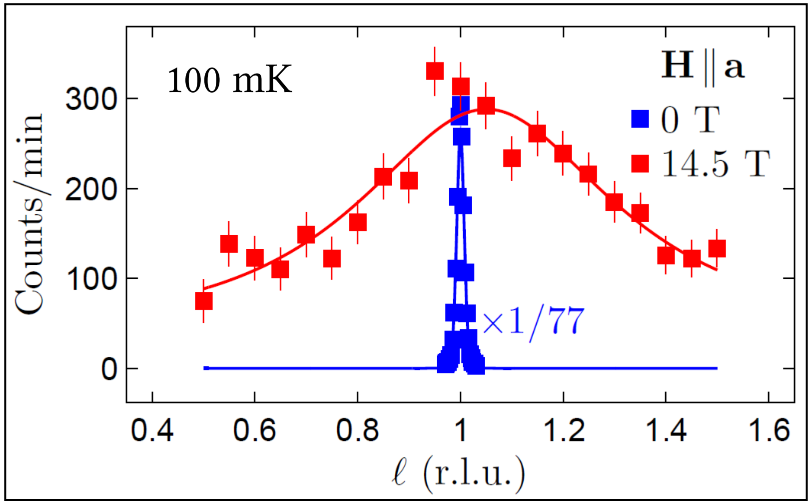

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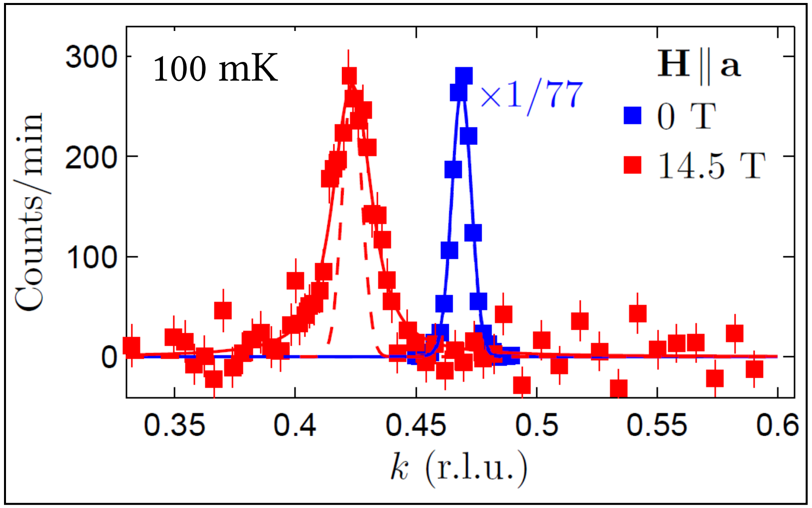

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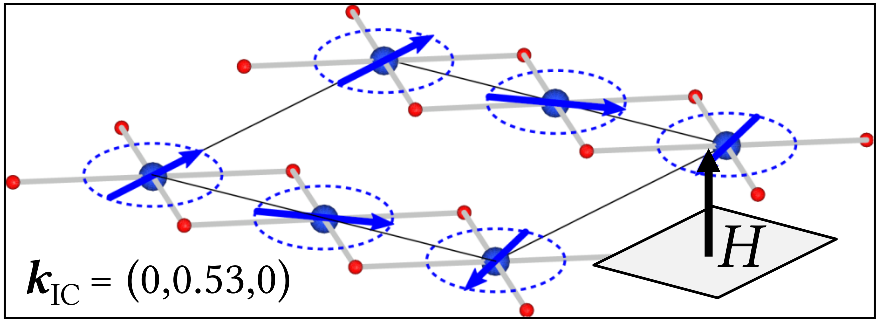

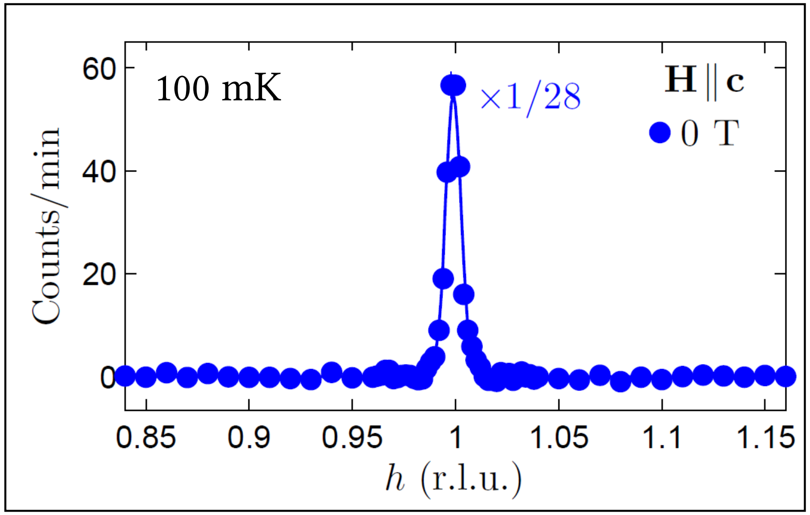

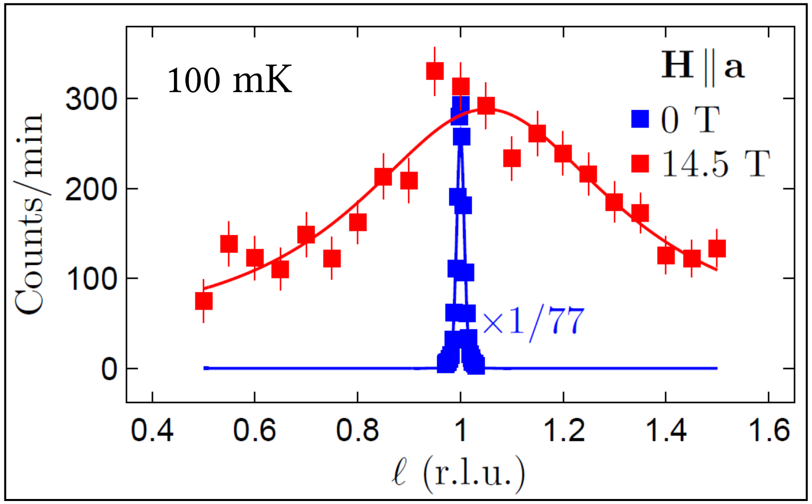

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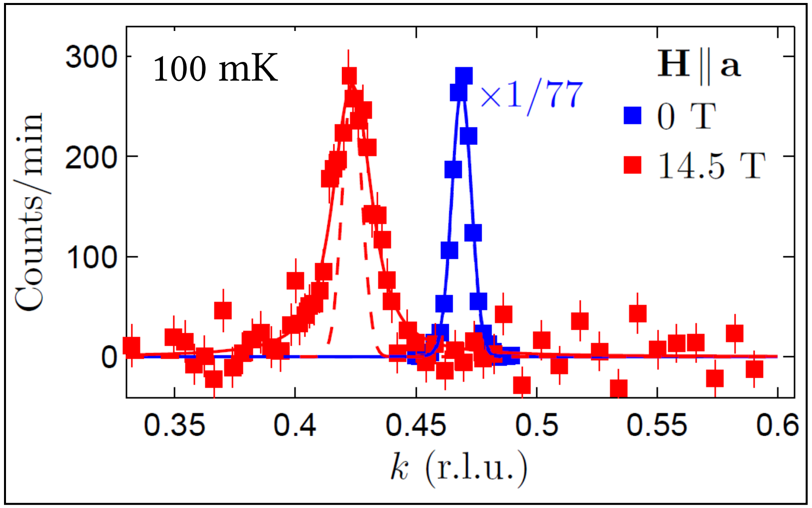

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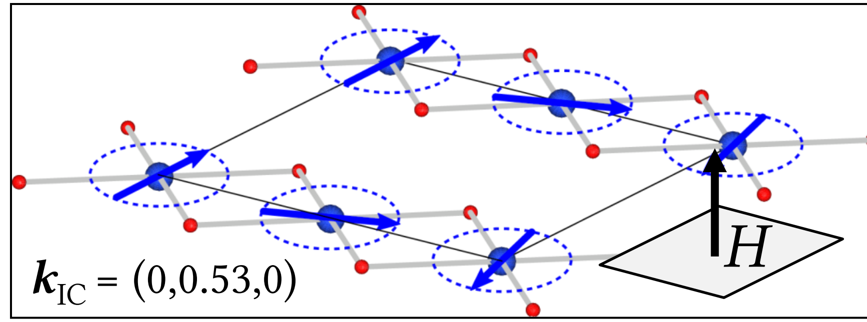

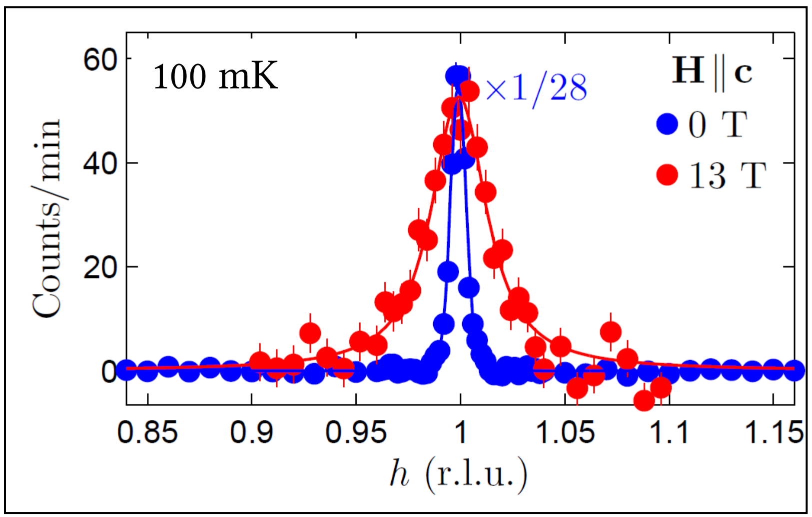

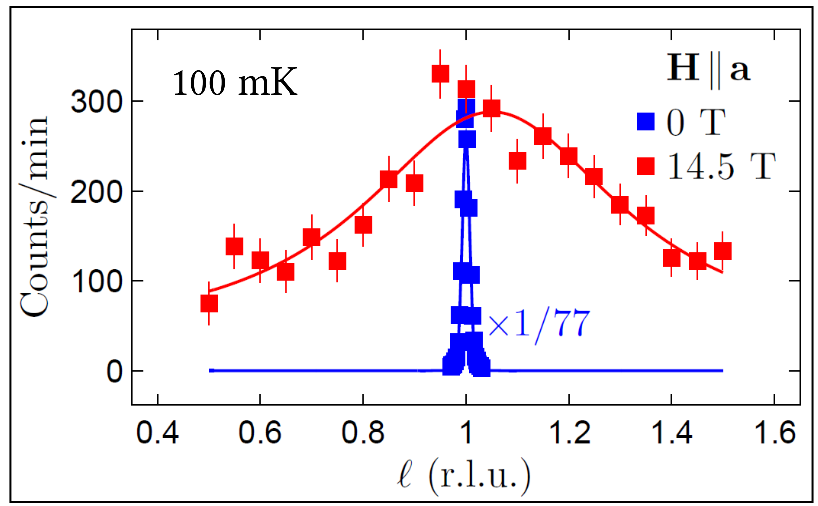

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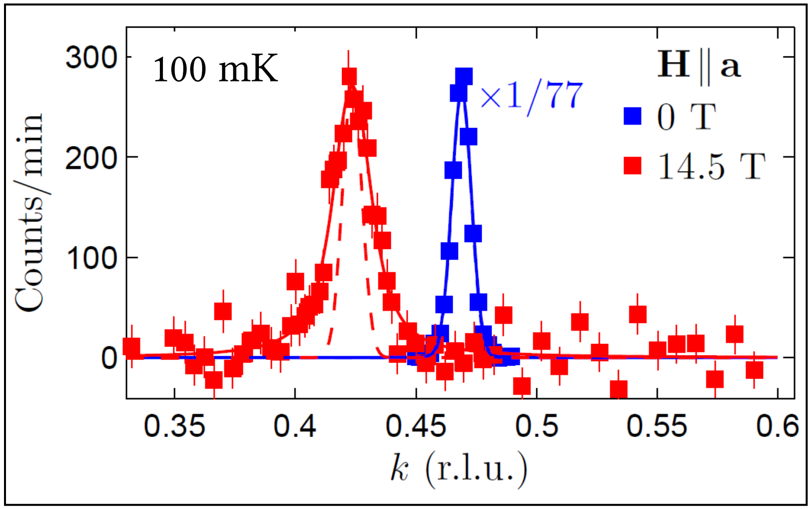
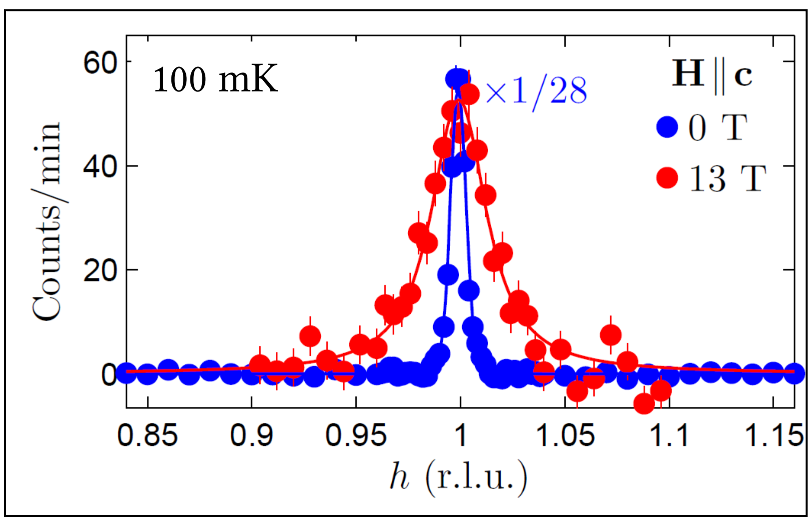
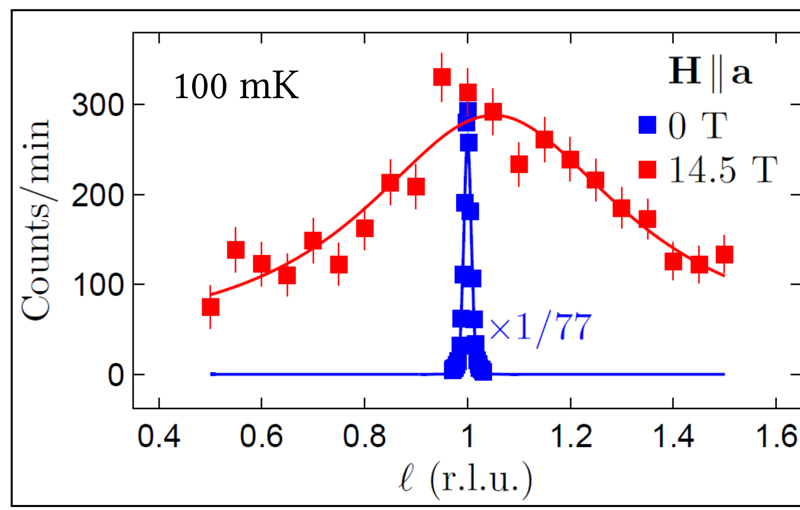


Dipolar correlations are short-range in all directions above H_Q at 100 mK

Integrated intensity is conserved





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2.2 Above H_Q=8T

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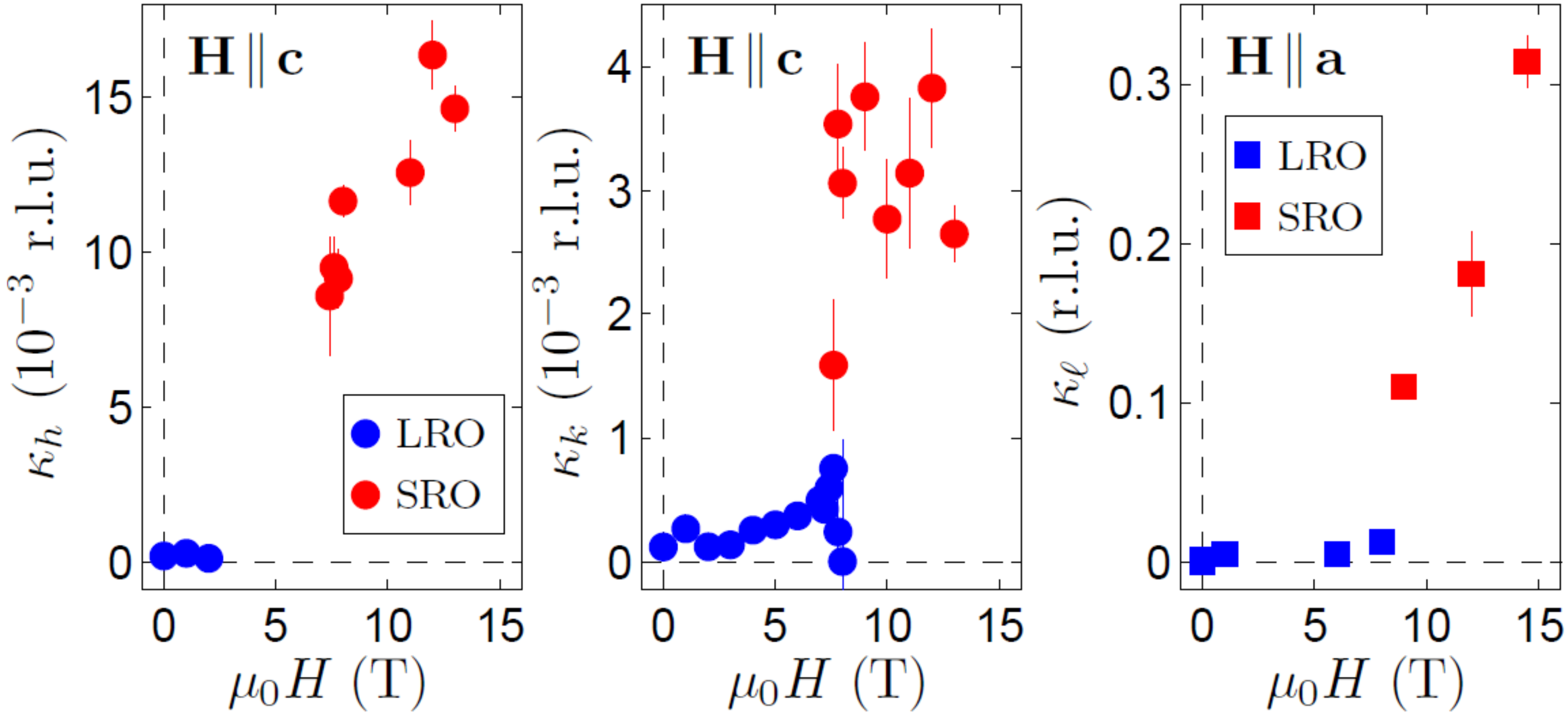
Abrupt broadening at H_Q

$\xi_a \sim 70$ nn, $\xi_b \sim 700$ nn, $\xi_c \sim 6$ nn

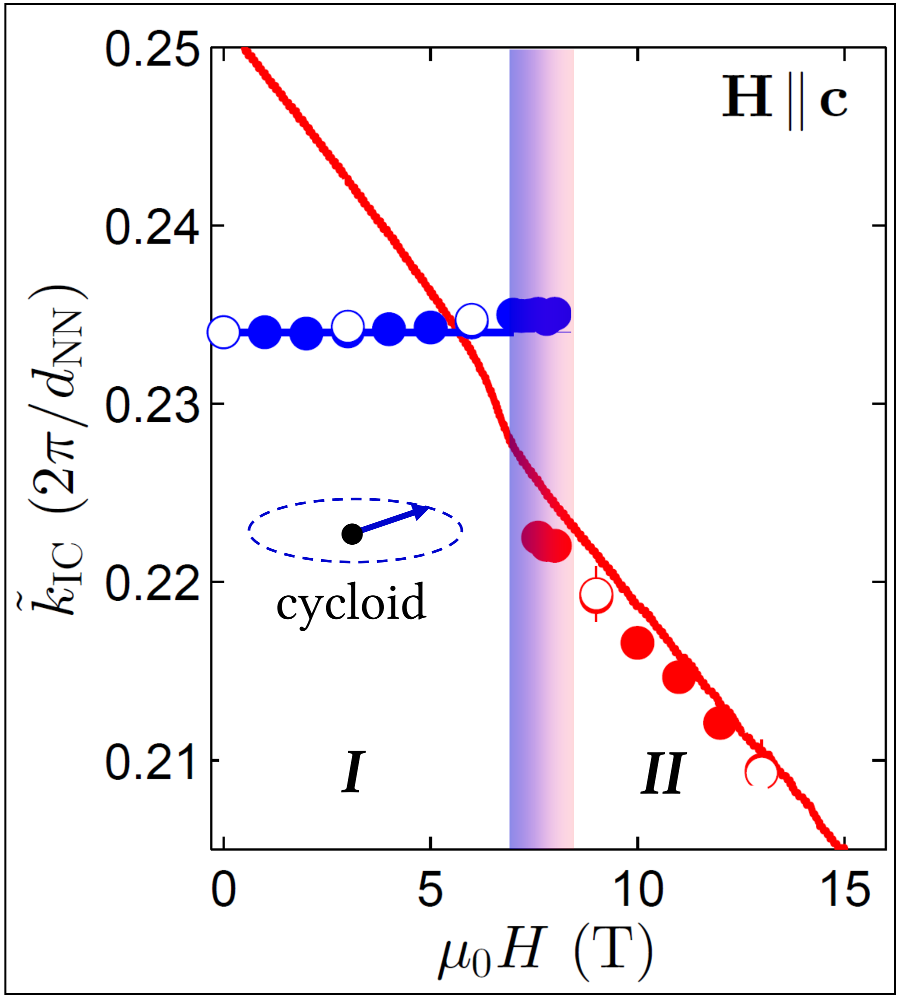
Dipolar correlations are short-range in all directions above H_Q at 100 mK

Integrated intensity is conserved



2.2 Above H_Q=8T

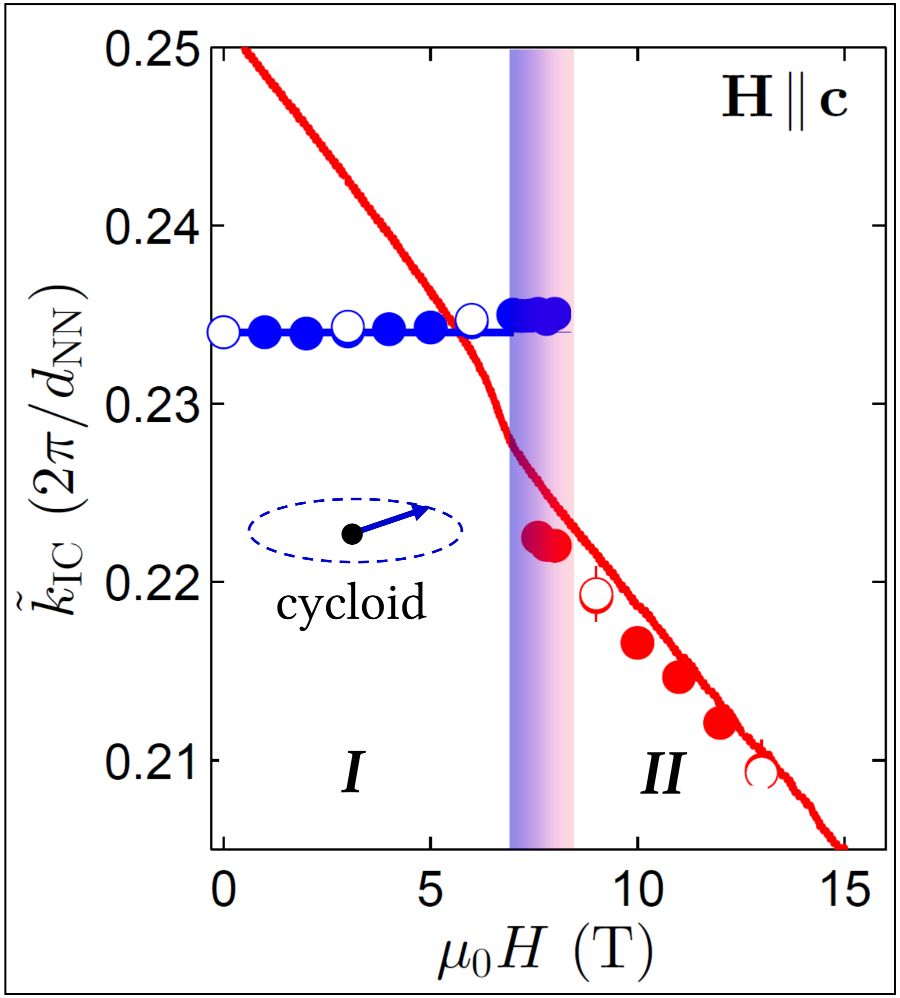
❖ 2. Field-dependence of dipolar correlations



See: Vekua *et al.*, PRB'07, Sato *et al.*, PRB'09. Magnetization curve: Svistov *et al.*, JETP Letters '11 T = 1.3 K

2.2 Above H_Q=8T

❖ 2. Field-dependence of dipolar correlations



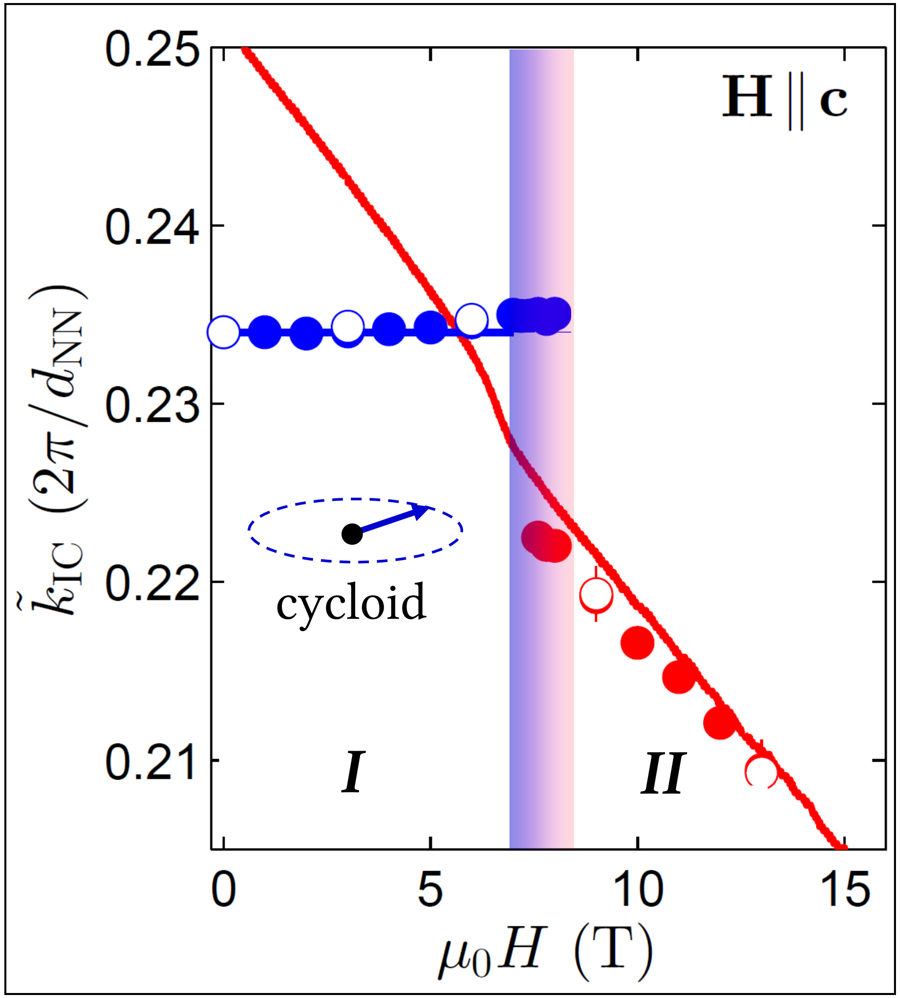
LR $\tilde{k}_{IC} = 1/4 - \delta$

SR $\tilde{k}_{IC} = 1/4 - m/2$

See: Vekua *et al.*, PRB'07, Sato *et al.*, PRB'09. Magnetization curve: Svistov *et al.*, JETP Letters '11 T = 1.3 K

2.2 Above H_Q=8T

❖ 2. Field-dependence of dipolar correlations



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

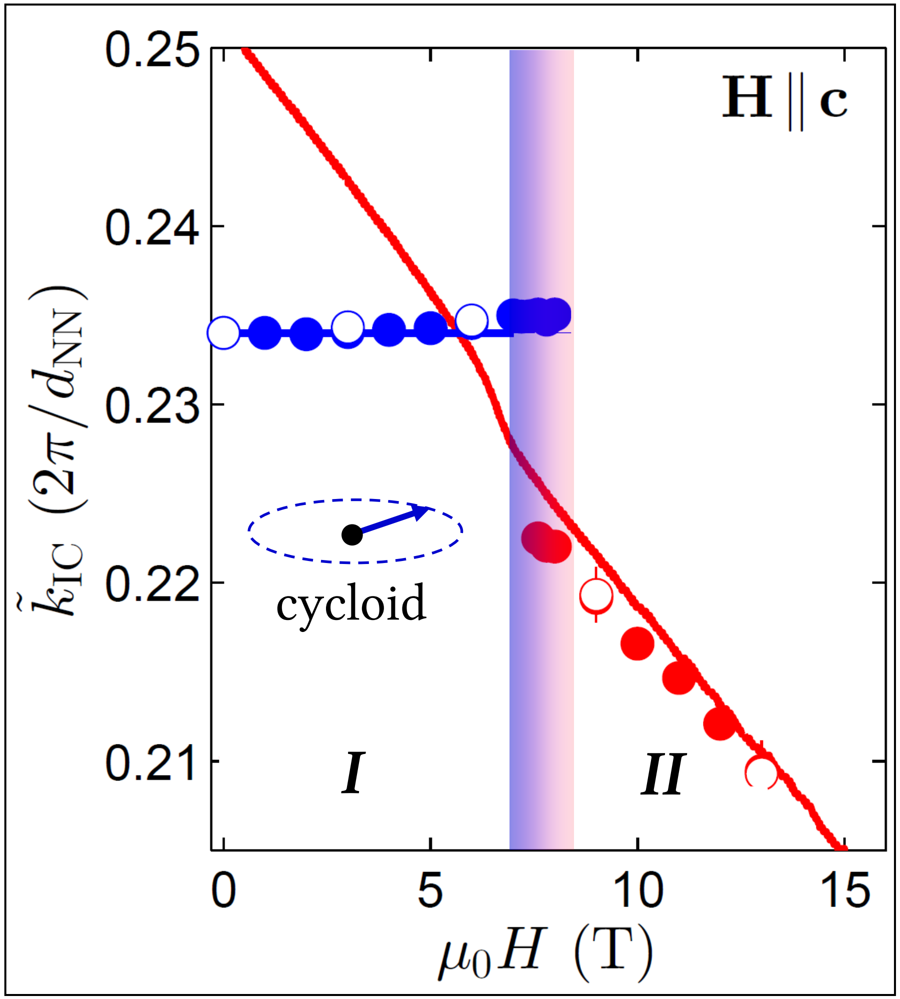
SR $\tilde{k}_{IC} = 1/4 - m/2$

Magnon-pairs in the ground-state

See: Vekua *et al.*, PRB'07, Sato *et al.*, PRB'09. Magnetization curve: Svistov *et al.*, JETP Letters '11 T = 1.3 K

2.2 Above H_Q=8T

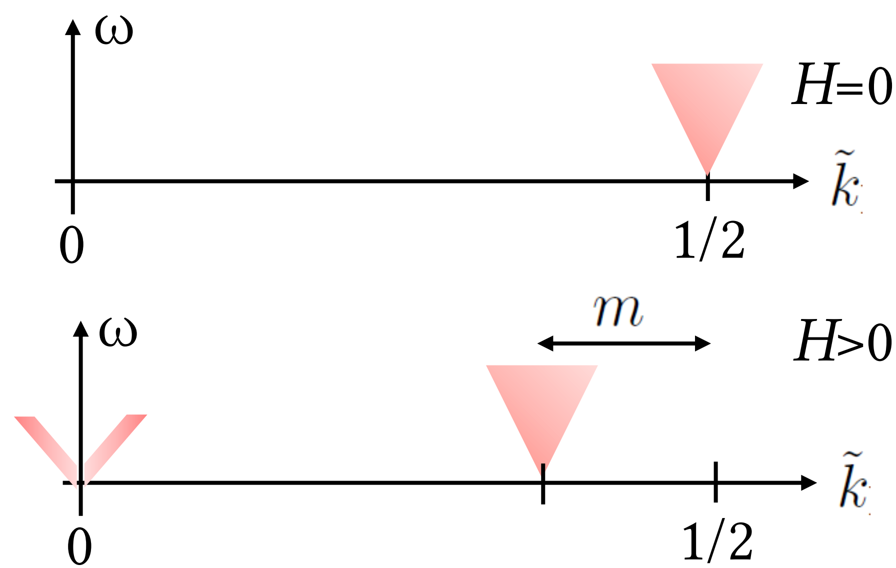
❖ 2. Field-dependence of dipolar correlations



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

SR $\tilde{k}_{IC} = 1/4 - m/2$

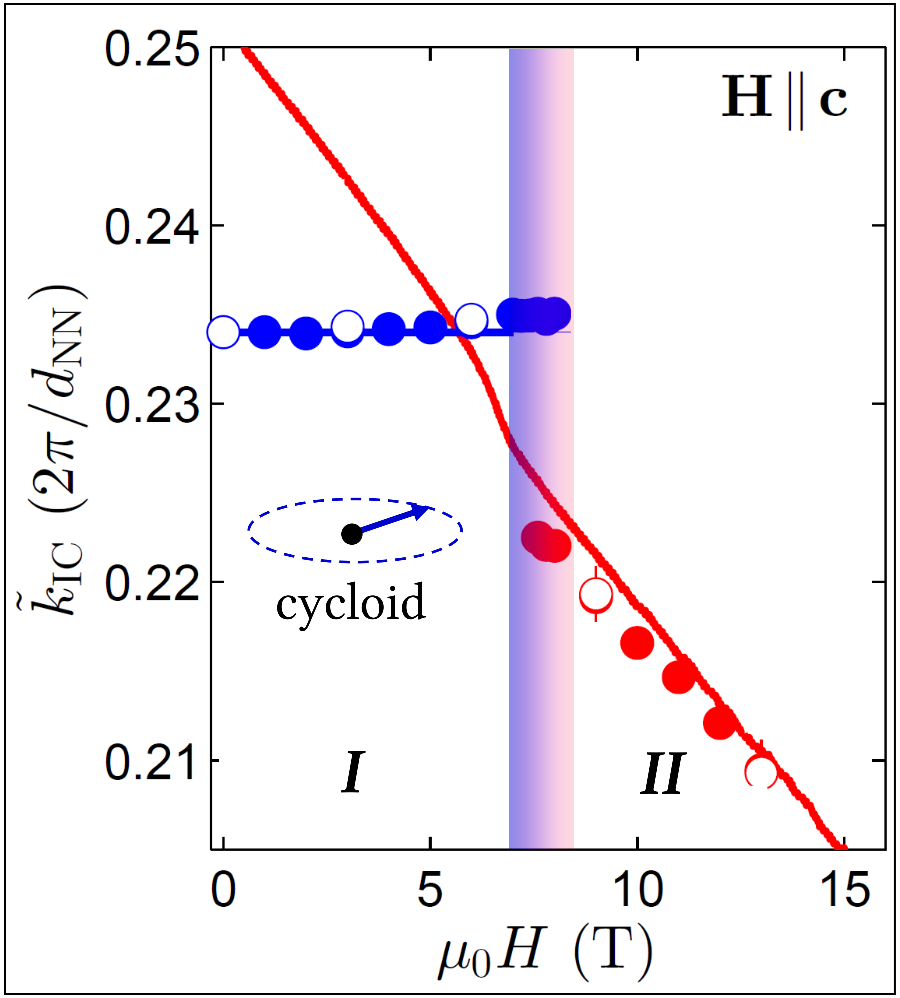
S^{zz} for a 1D chain

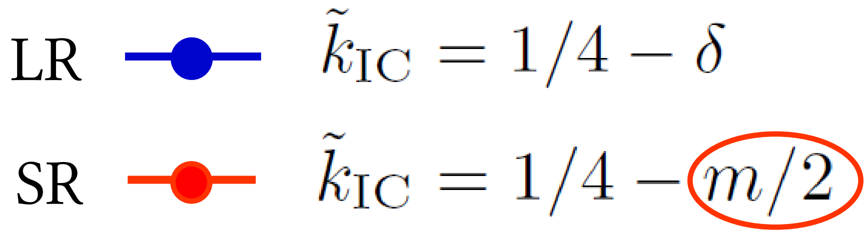


See: Vekua *et al.*, PRB'07, Sato *et al.*, PRB'09. Magnetization curve: Svistov *et al.*, JETP Letters '11 T = 1.3 K

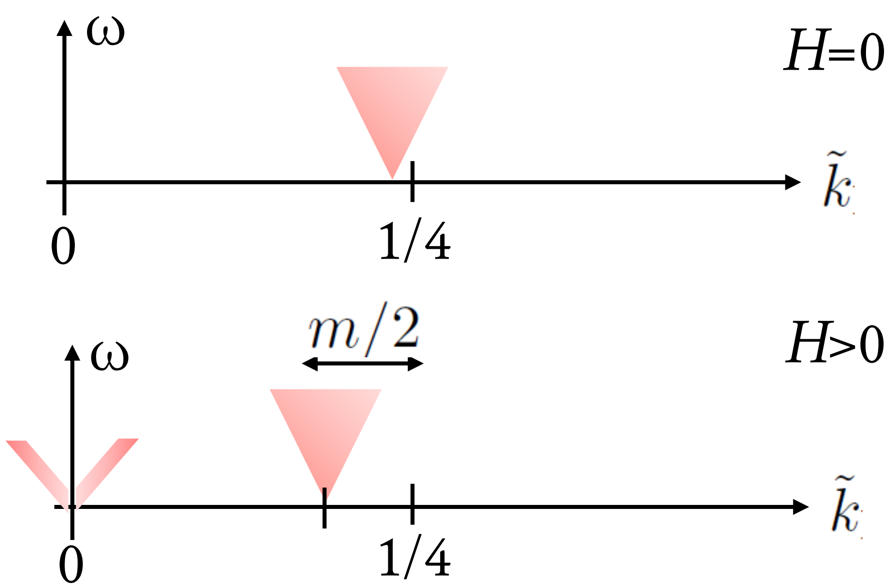
2.2 Above H_Q=8T

❖ 2. Field-dependence of dipolar correlations





S^{zz} in the quadrupolar-nematic phase

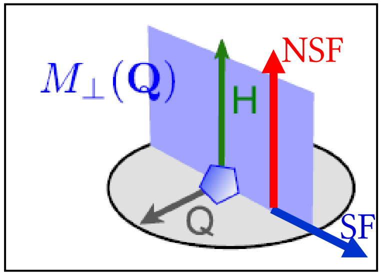


See: Vekua *et al.*, PRB'07, Sato *et al.*, PRB'09. Magnetization curve: Svistov *et al.*, JETP Letters '11 T = 1.3 K

2.2 Above H_Q=8T

❖ 3. Spin components involved in short-range correlations

Polarized neutrons, vertical field, 50 mK



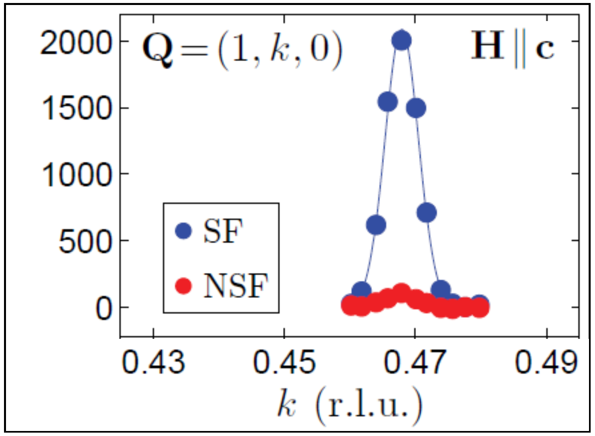
$\boxed{\sigma_{\mathrm{SF}},\ \perp \mathbf{H} \perp \mathbf{Q}}$ In-plane

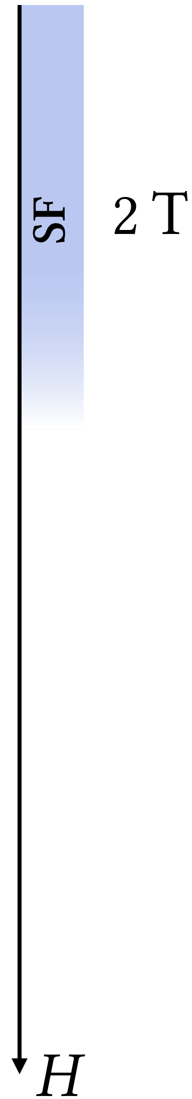
$\boxed{\sigma_{\mathrm{NSF}},\ || \mathbf{H} \perp \mathbf{Q}}$ Out-of-plane

Instrument: IN14, ILL, Grenoble with 12T asymmetric magnet

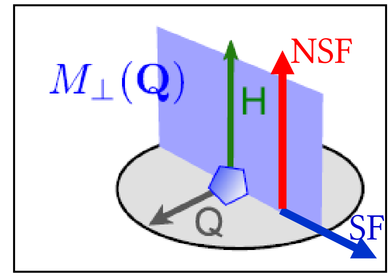
2.2 Above H_Q=8T

❖ 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

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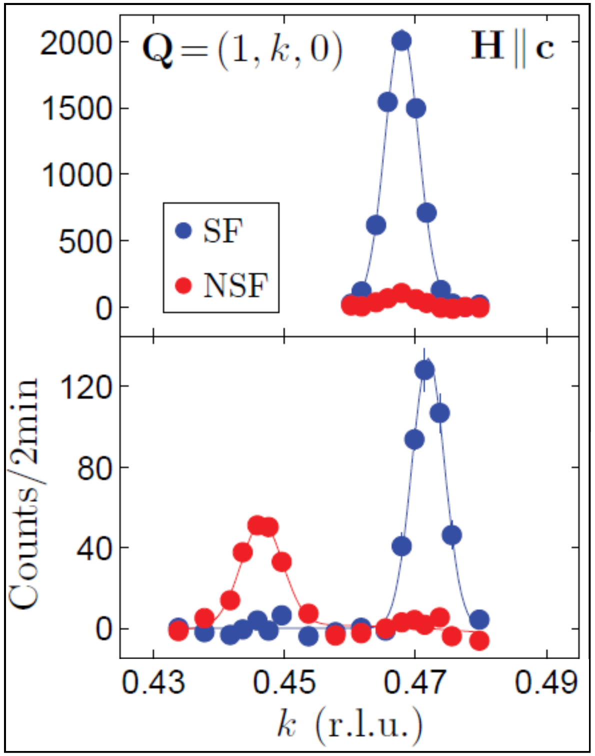
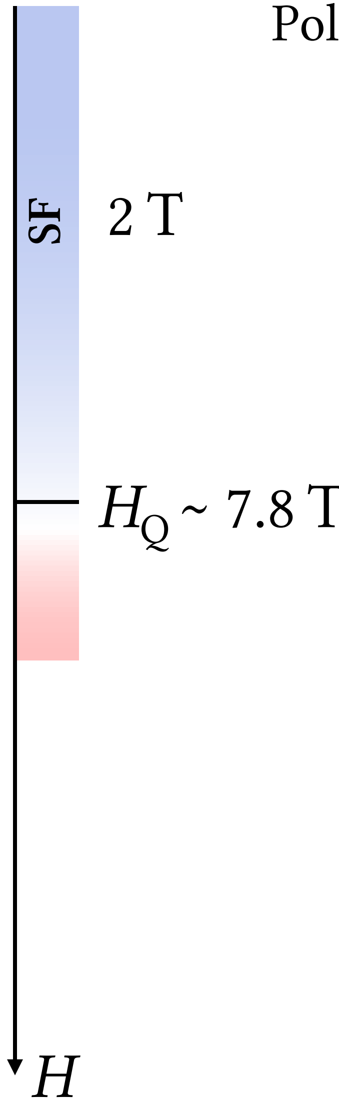
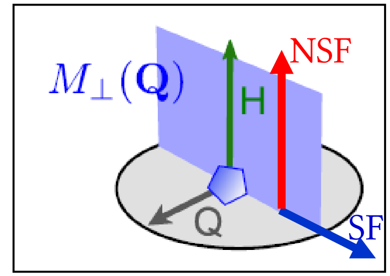
2.2 Above H_Q=8T

❖ 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

Instrument: IN14, ILL, Grenoble with 12T asymmetric magnet

2.2 Above H_Q=8T

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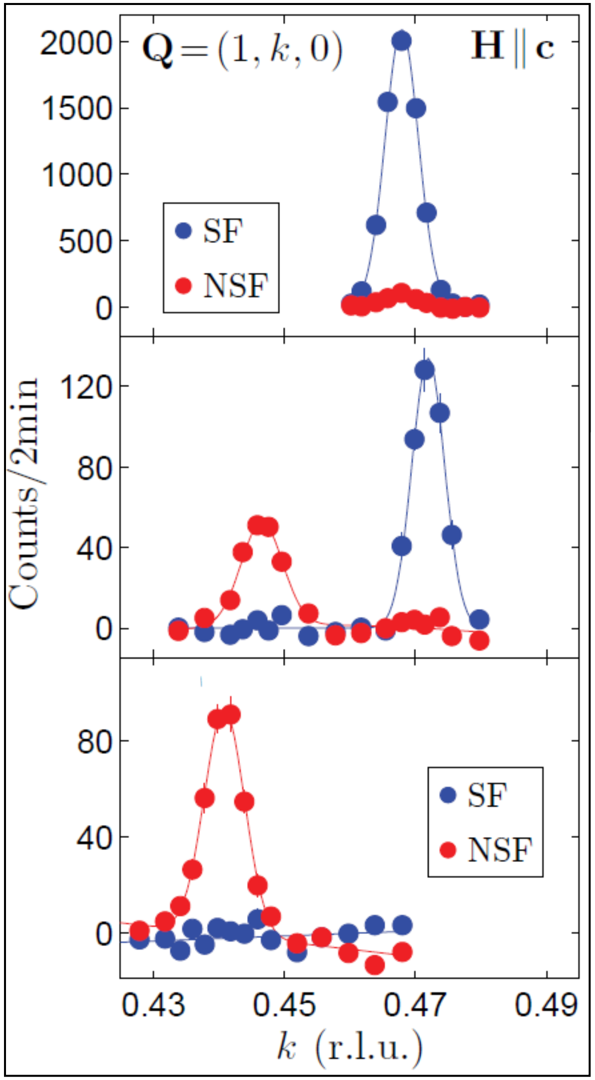
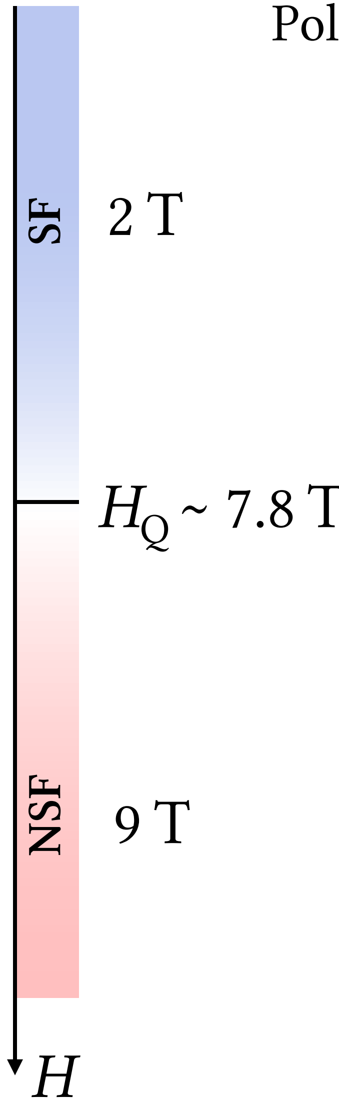
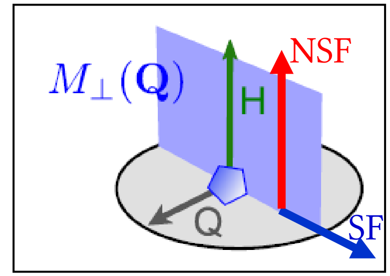




Polarized neutrons, vertical field, 50 mK



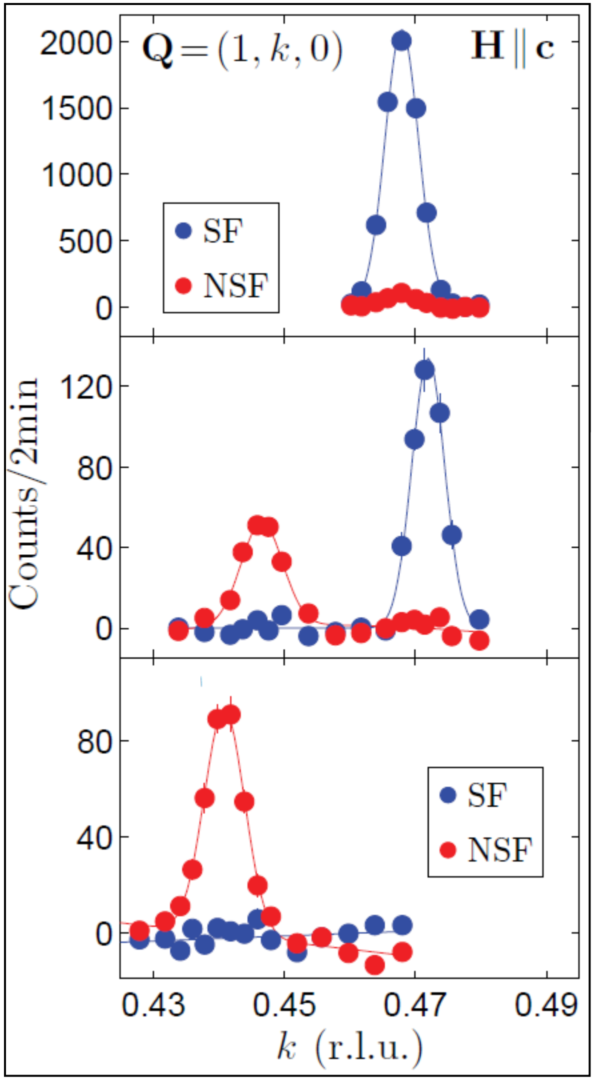
$\boxed{\sigma_{\mathrm{SF}}, \perp \mathbf{H} \perp \mathbf{Q}}$ In-plane

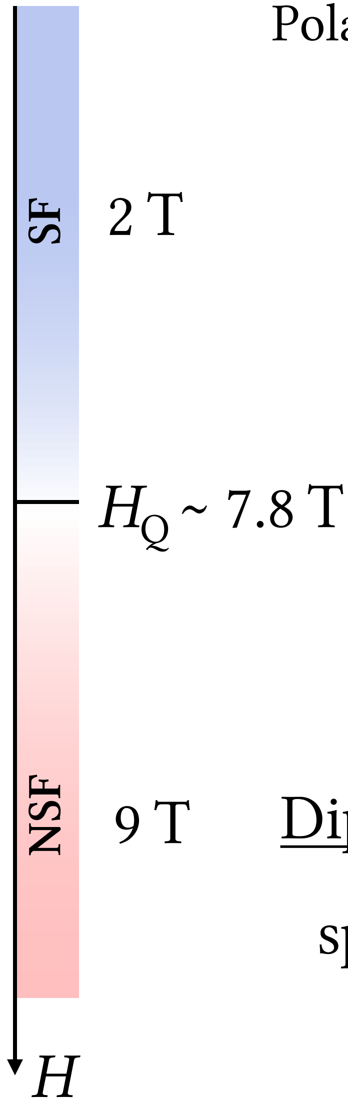
$\boxed{\sigma_{\mathrm{NSF}}, \parallel \mathbf{H} \perp \mathbf{Q}}$ Out-of-plane

Instrument: IN14, ILL, Grenoble with 12T asymmetric magnet

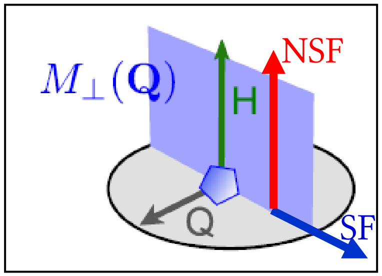
2.2 Above H_Q=8T

❖ 3. Spin components involved in short-range correlations





Polarized neutrons, vertical field, 50 mK



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

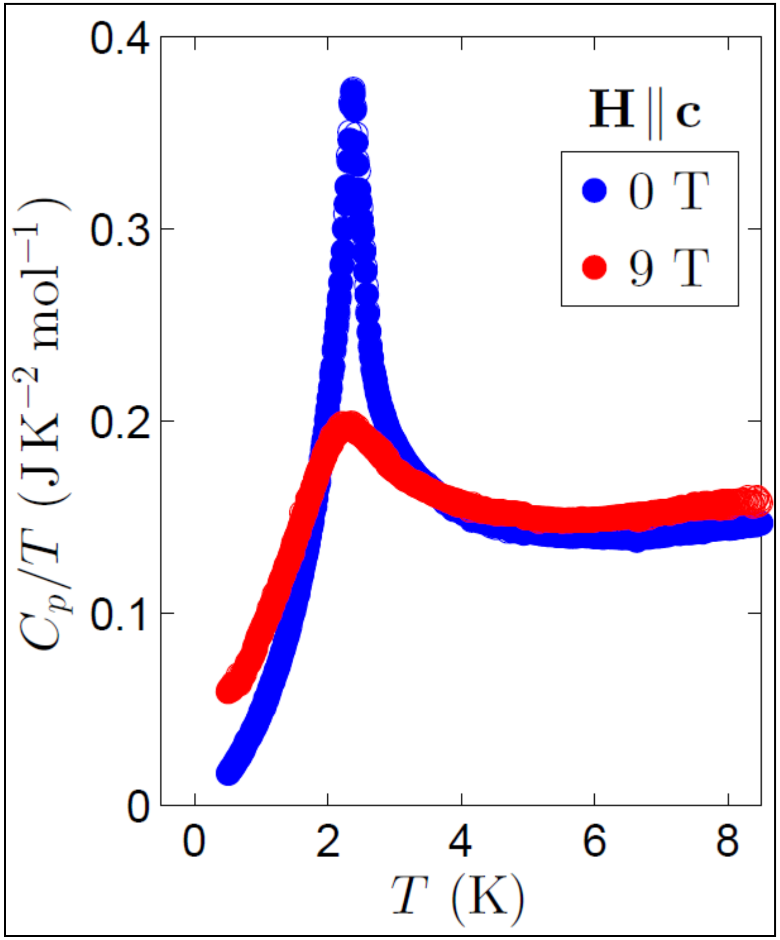
$\boxed{\sigma_{NSF}, \parallel \mathbf{H} \perp \mathbf{Q}}$ Out-of-plane

Dipolar short-range involve only spin components parallel to **H**

Instrument: IN14, ILL, Grenoble with 12T asymmetric magnet

2.2 Above H_Q=8T

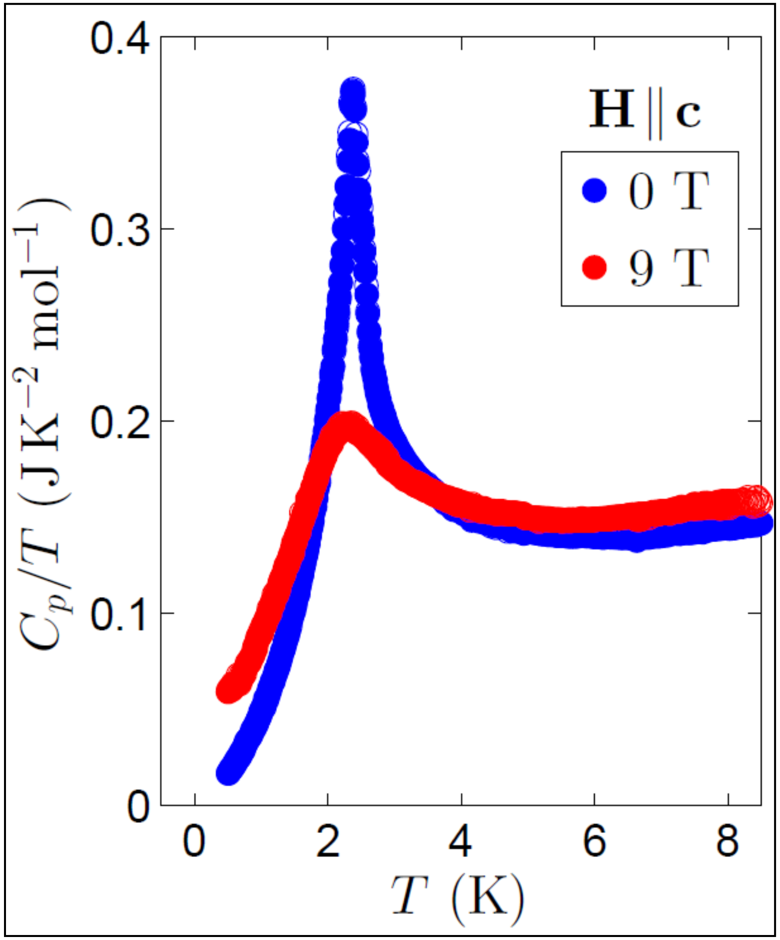
❖ 4. Phase-transition evidenced above H_Q

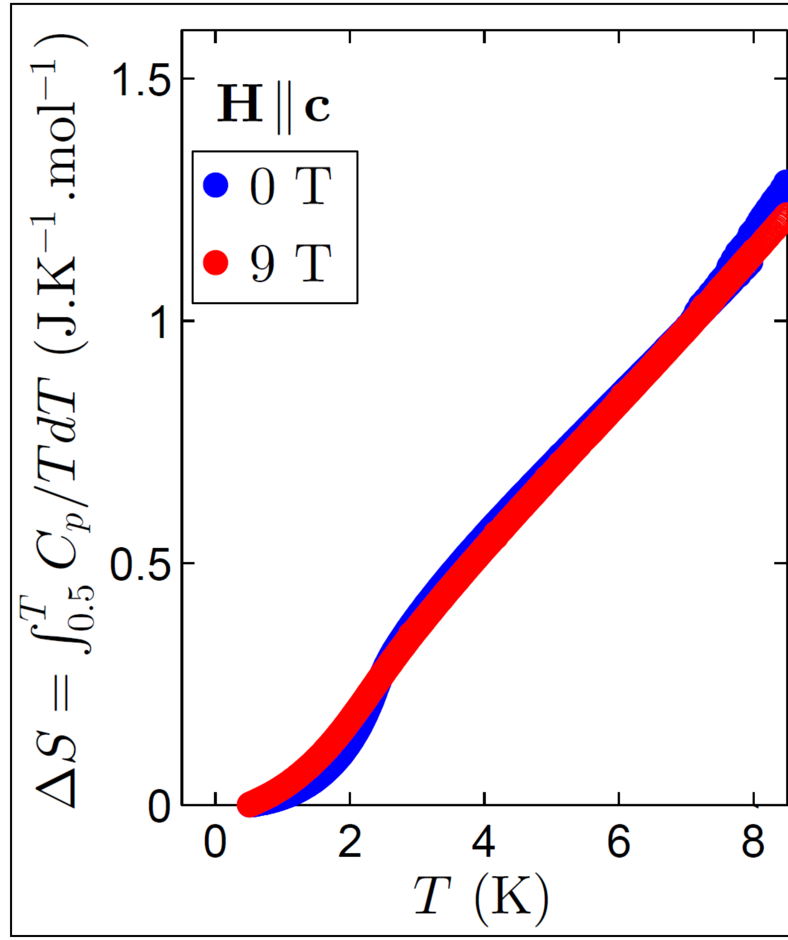


Measurement: J. M. Law and R. K. Kremer, MPI Stuttgart

2.2 Above H_Q=8T

❖ 4. Phase-transition evidenced above H_Q

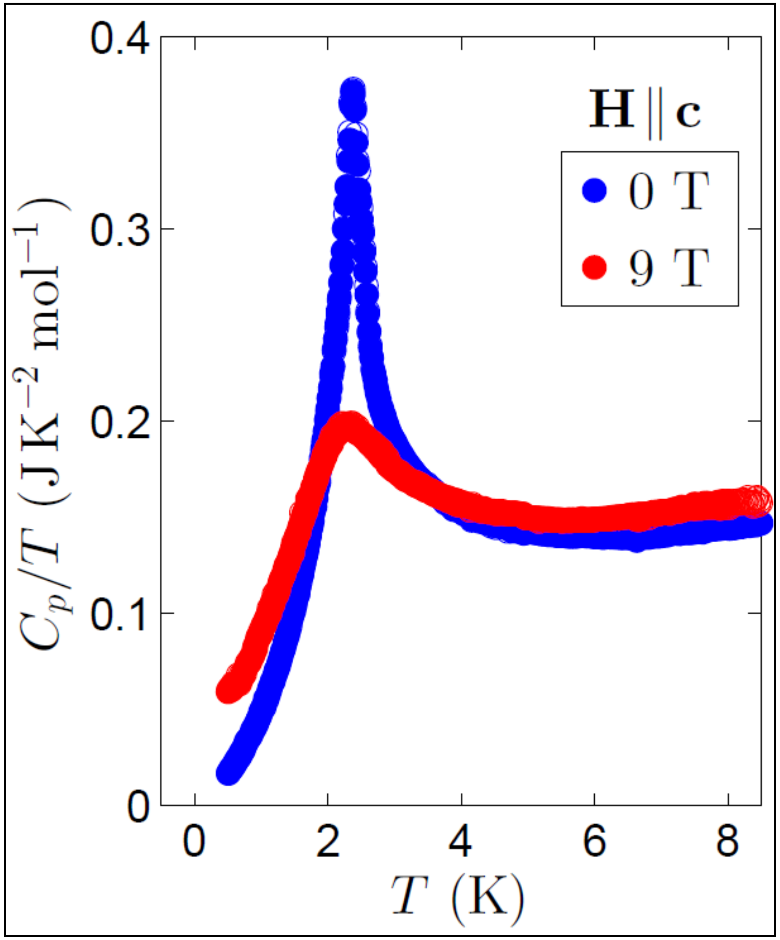


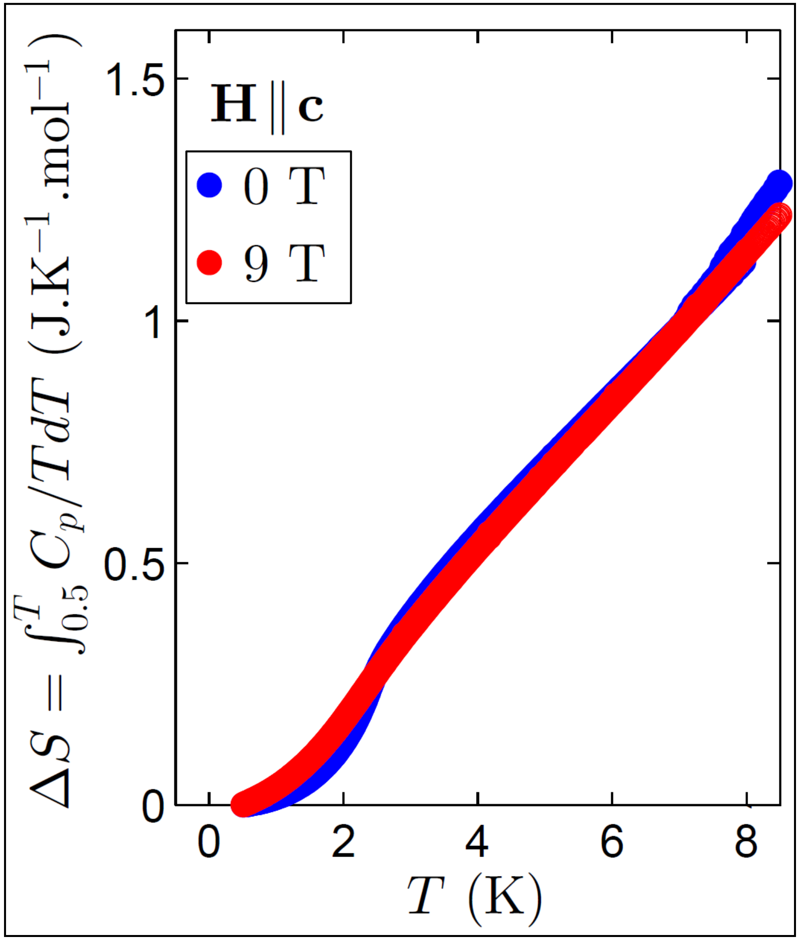


<u>Measurement:</u> J. M. Law and R. K. Kremer, MPI Stuttgart

2.2 Above H_Q=8T

❖ 4. Phase-transition evidenced above H_Q





Indication for a thermal phase transition above H_Q

Measurement: J. M. Law and R. K. Kremer, MPI Stuttgart

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**

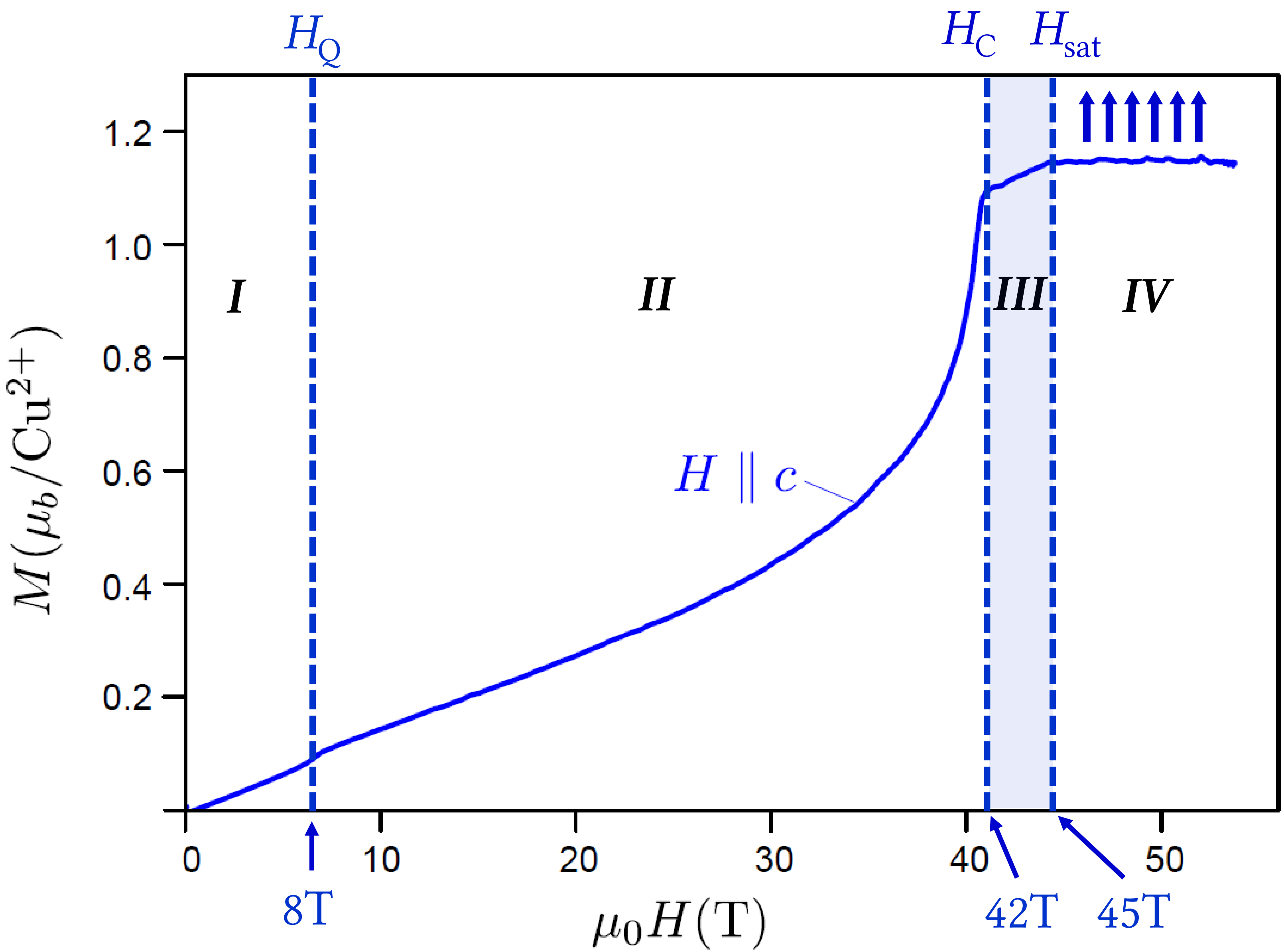
1. Short-range dipolar correlations in all directions

2. Driven by quadrupolar-nematic correlations

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

4. Thermal phase transition

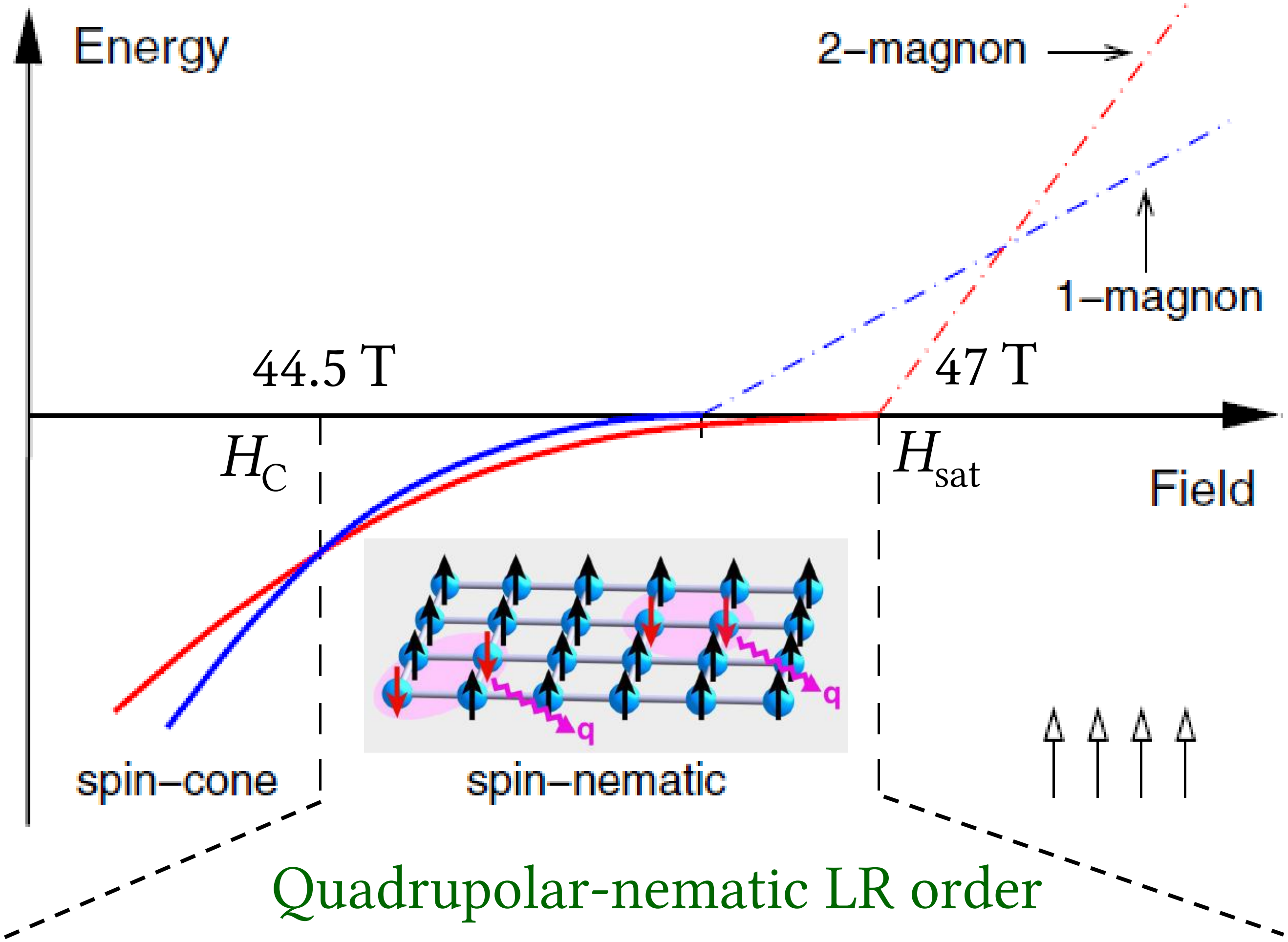
2.3 Magnetization curve of $LiCuVO_4$



<u>See</u>: Banks *et al.*, JPCM '07, Svistov *et al.*, JETP Letters '11

2.3 Quadrupolar long-range order

❖ Quasi-1D system with frustrated interchain interactions

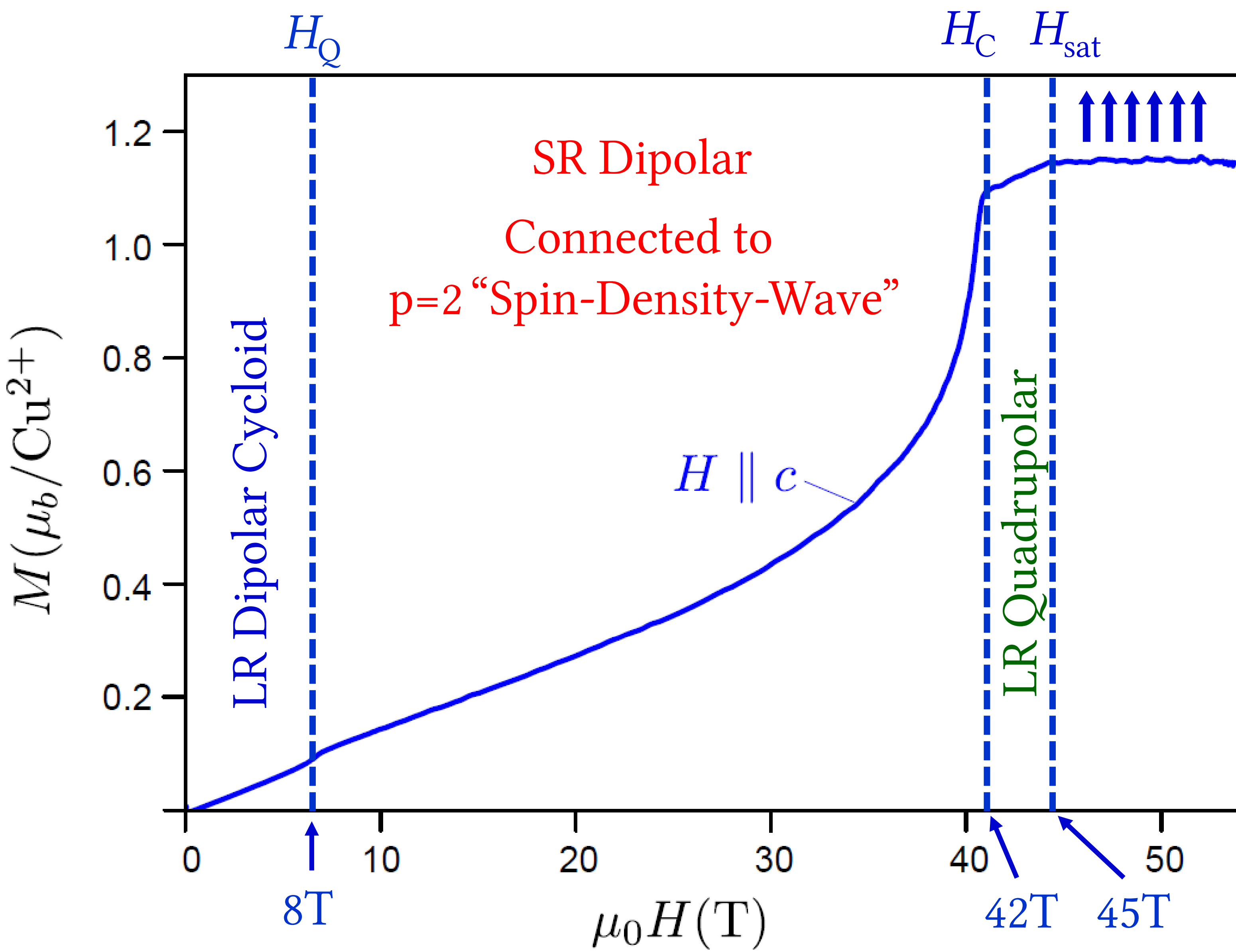


Quadrupolar-nematic LR order

SR transverse dipolar correlations

See: **Zhitomirsky and Tsunetsugu, EPL '10**

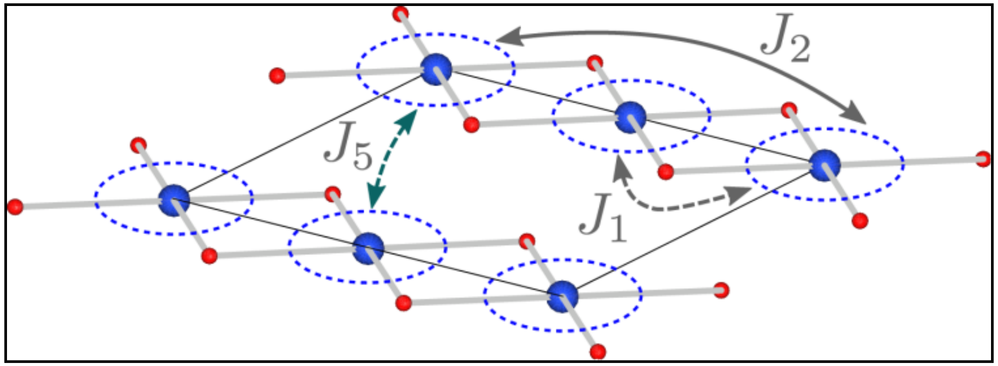
2.3 Magnetization curve of $LiCuVO_4$



See: Banks *et al.*, JPCM '07, Svistov *et al.*, JETP Letters '11

2.3 A Possible Scenario above H_Q=8T

❖ Role of frustrated inter-chain interactions



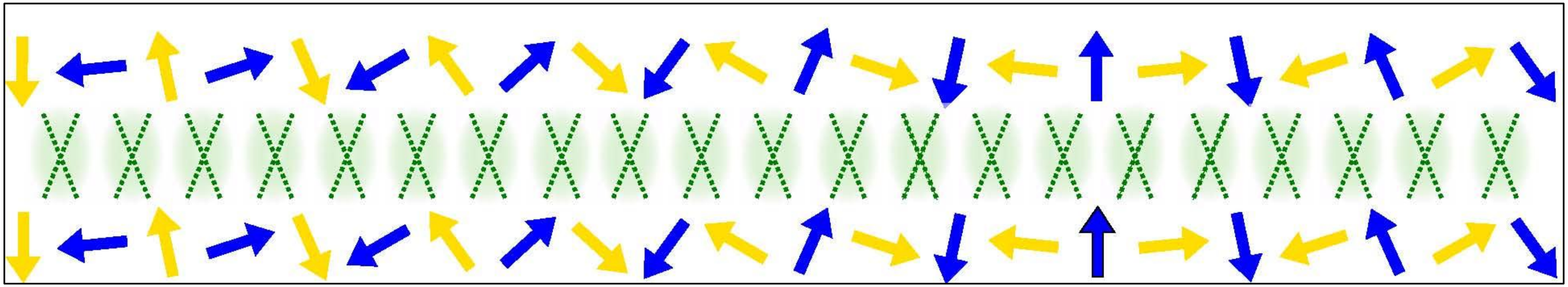
J_1 = -1.6 meV
J_2 = +5.6 meV
J_5 = -0.4 meV

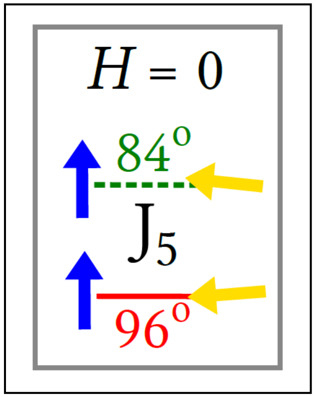
Enderle *et al.*, EPL '05

... Qualitative picture using solitons (fermions) ...

2.3 A Possible Scenario …

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





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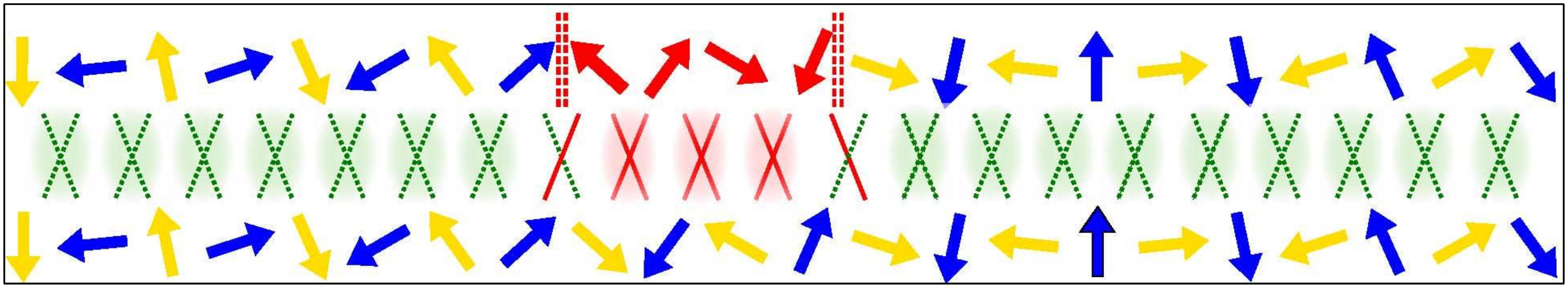
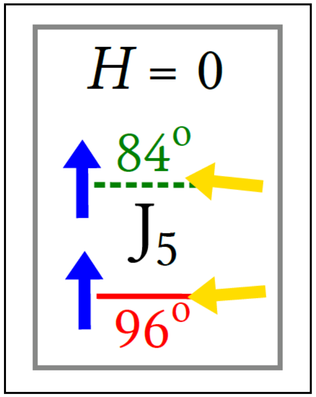


<u>Quantum Fluctuations:</u> 2-soliton + 2-soliton

Furukawa *et al.*, JPSJ '08



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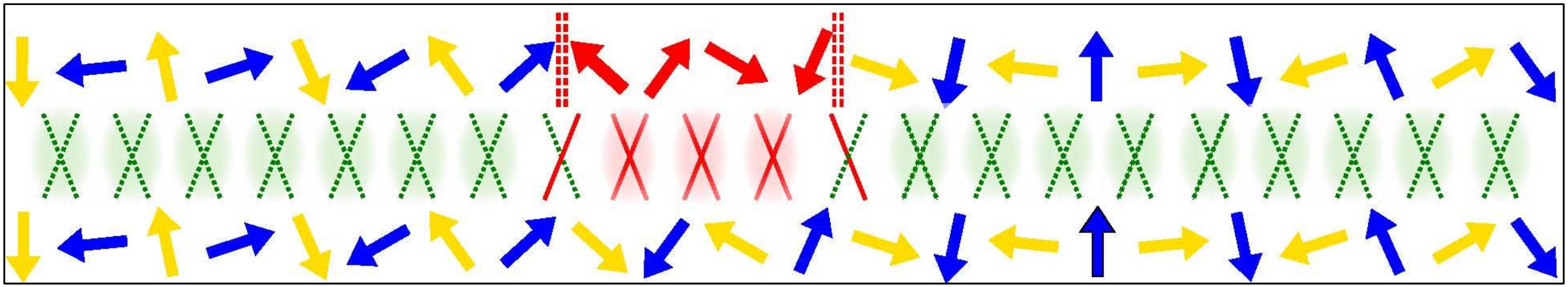
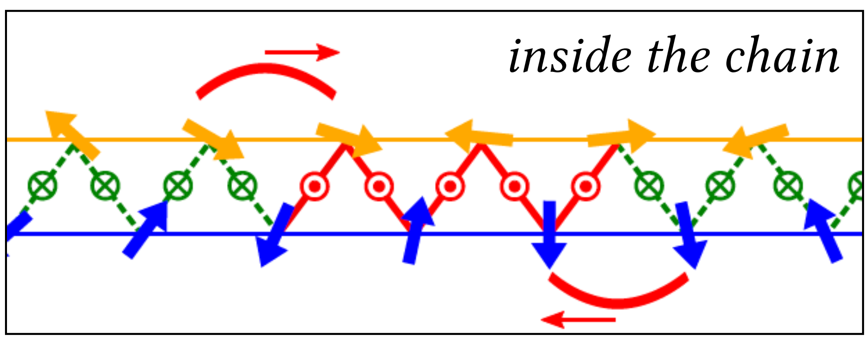
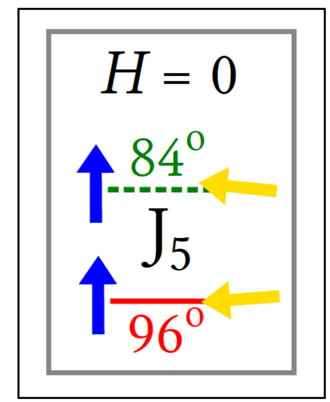


2-soliton + 2-soliton



A 2-soliton is bound together by FM J_1



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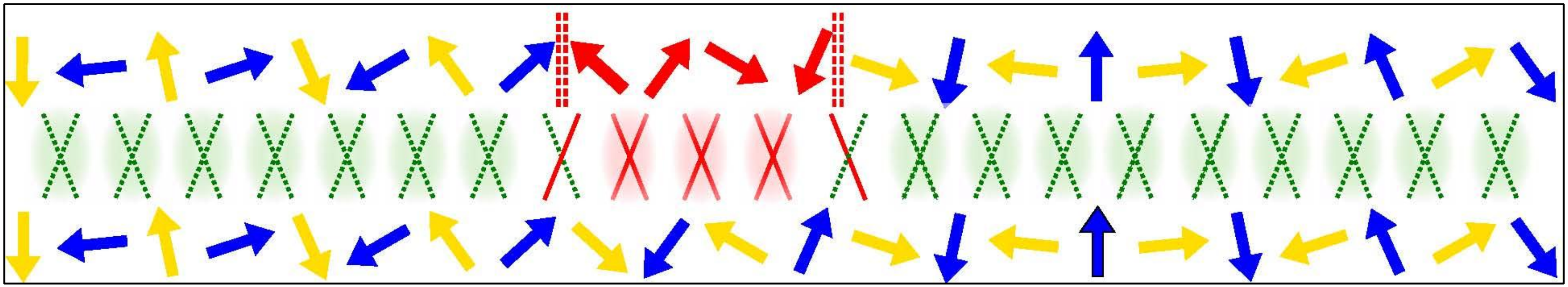
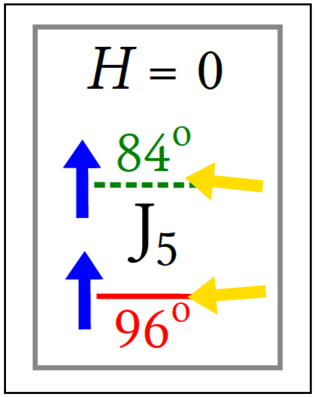
❖ Role of <u>frustrated</u> inter-chain interactions $H = 0$





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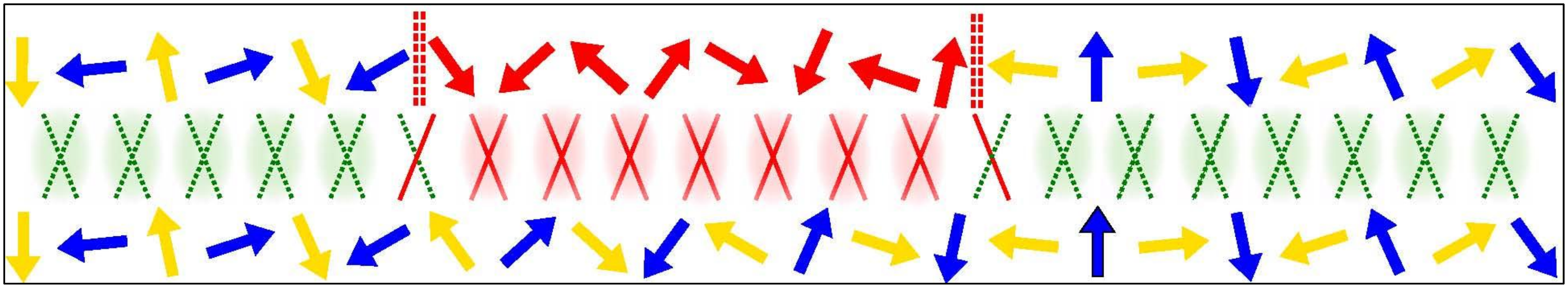
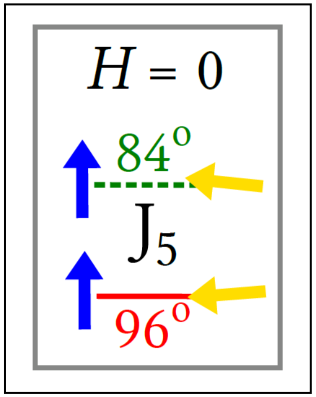
❖ Role of <u>frustrated</u> inter-chain interactions $H = 0$





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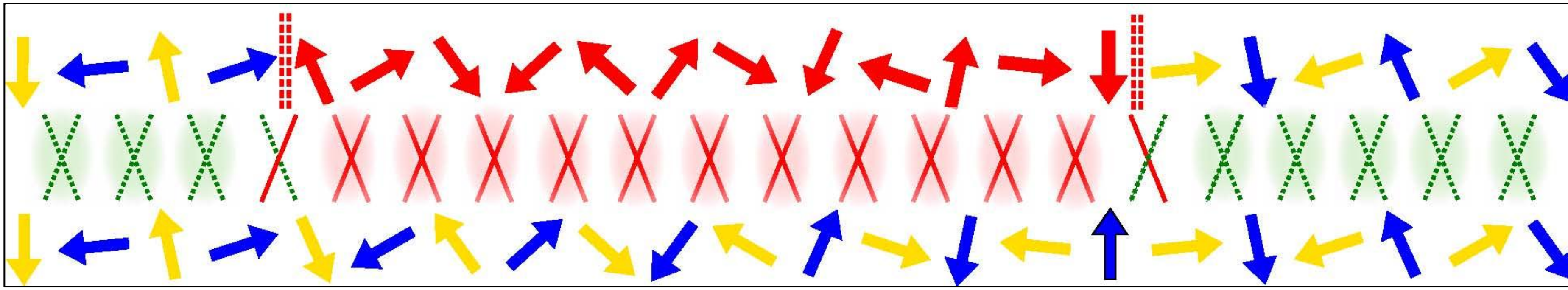

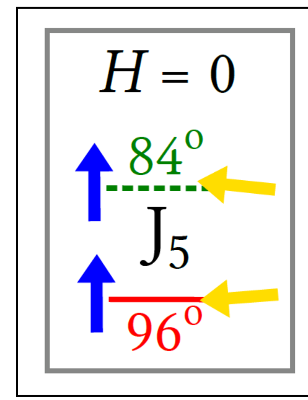

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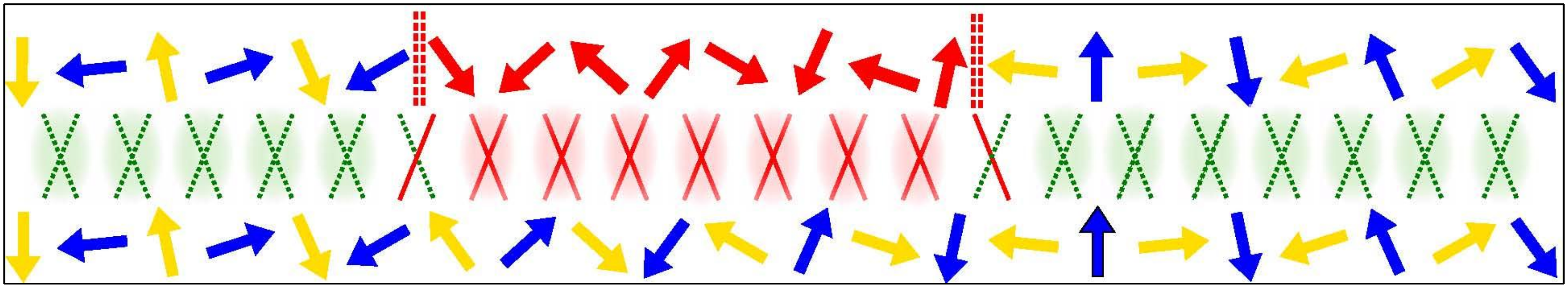
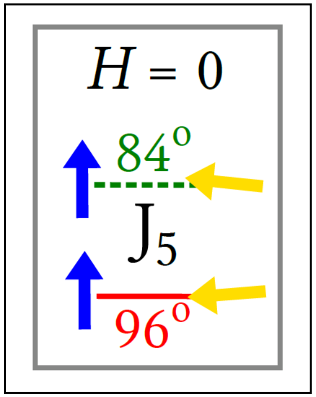
❖ Role of <u>frustrated</u> inter-chain interactions $H = 0$





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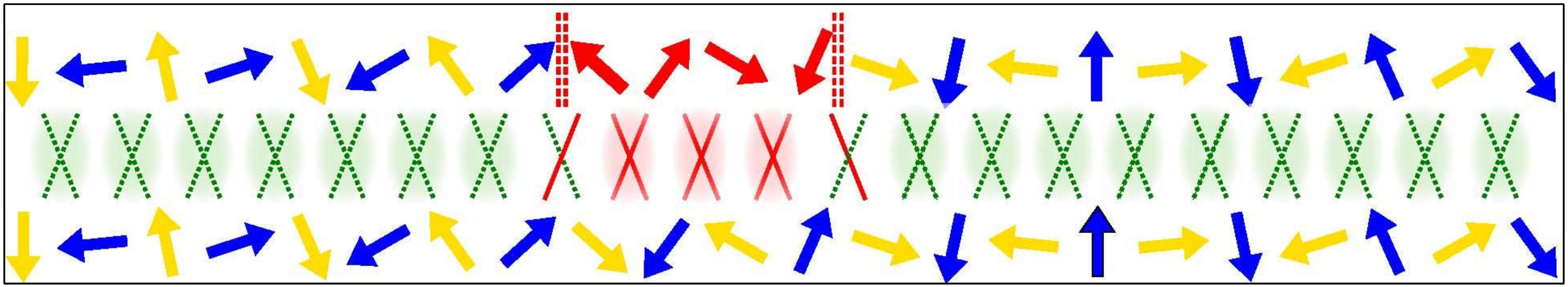
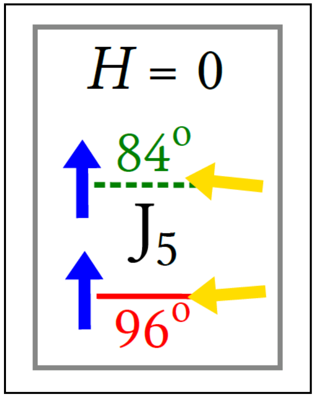
❖ Role of frustrated inter-chain interactions H = 0



Bound 4-soliton



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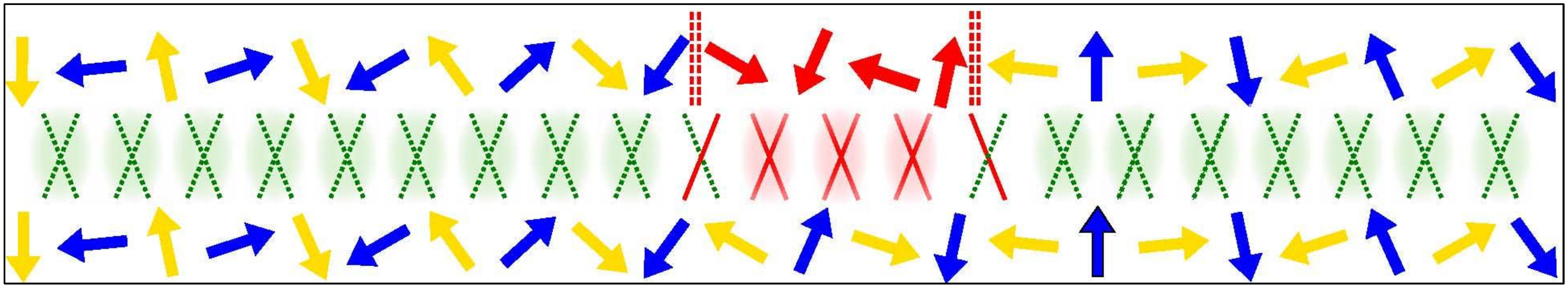
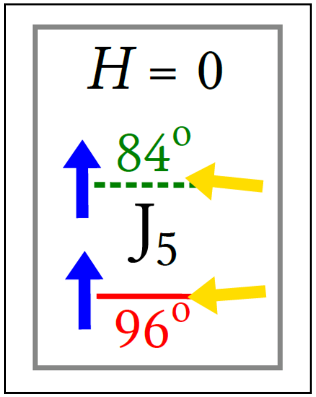
❖ Role of <u>frustrated</u> inter-chain interactions $H = 0$





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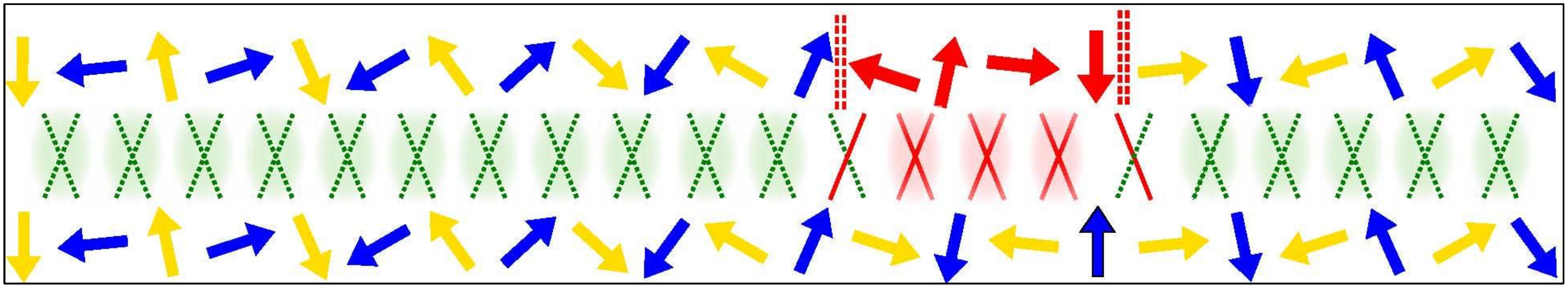

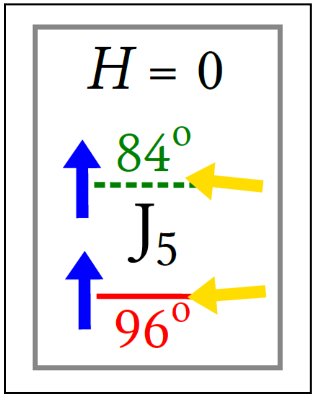

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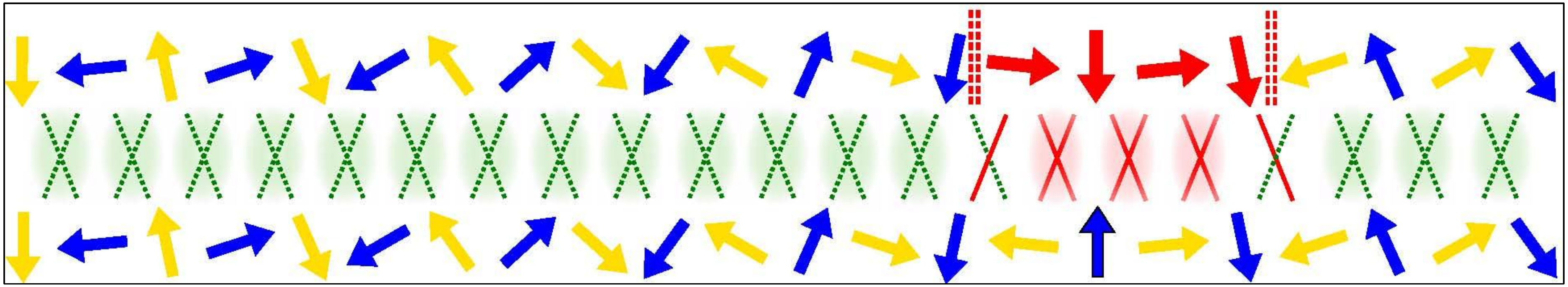
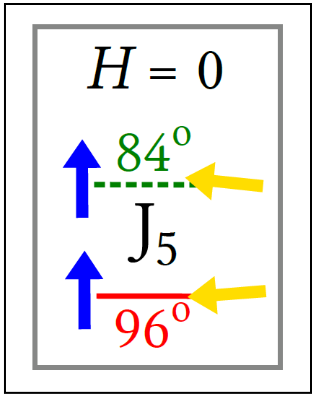
❖ Role of frustrated inter-chain interactions $H = 0$





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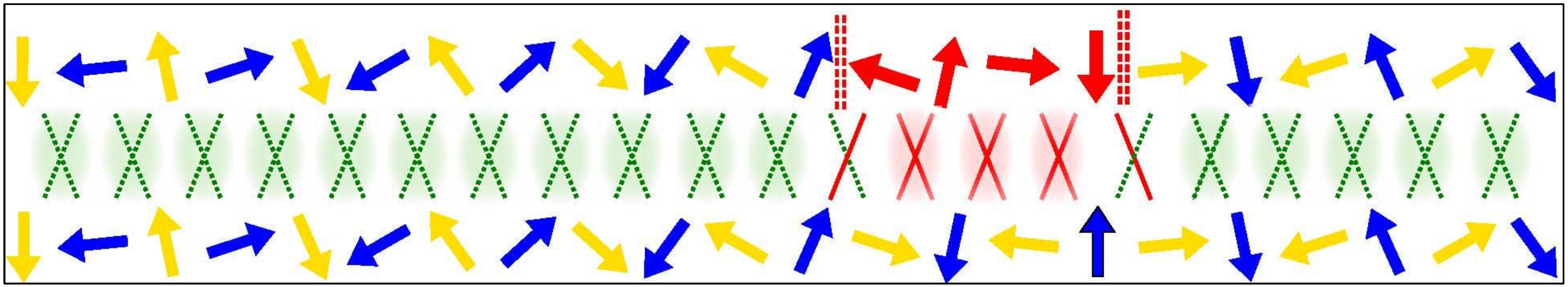
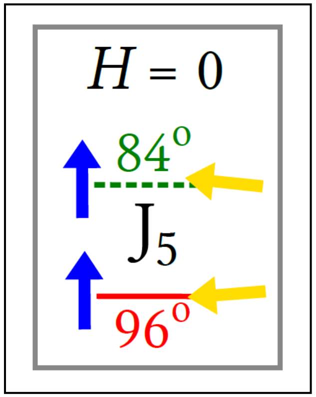
❖ Role of frustrated inter-chain interactions $H = 0$





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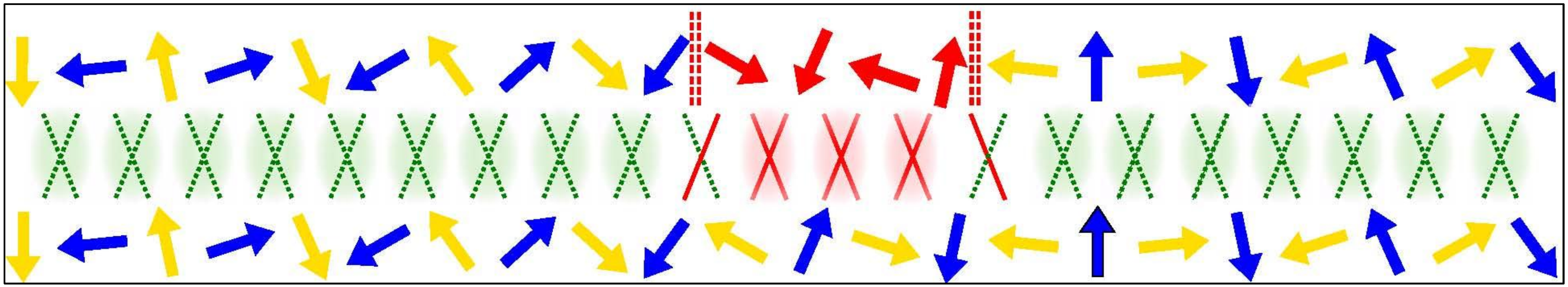
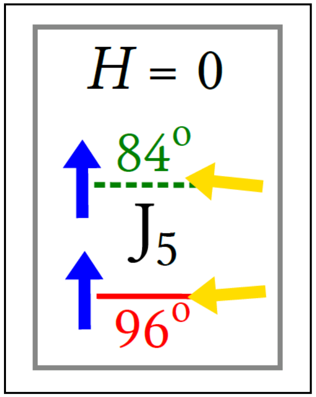
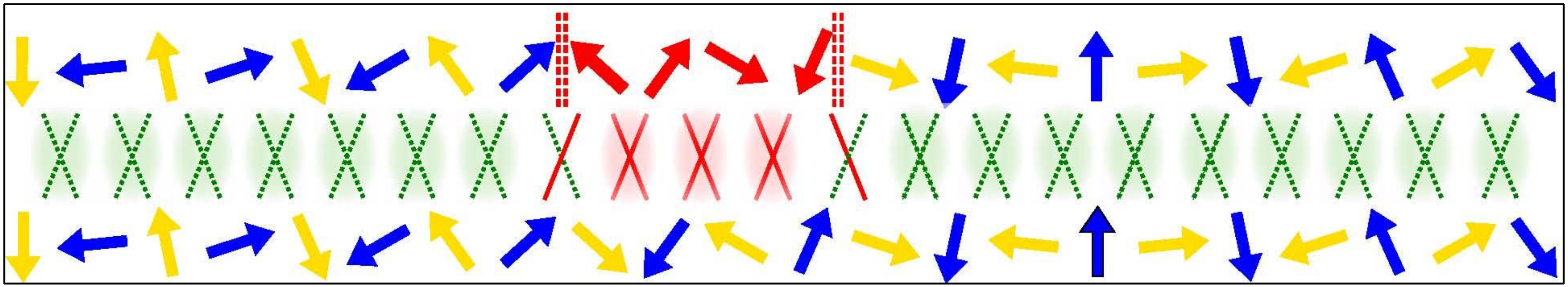
❖ Role of <u>frustrated</u> inter-chain interactions H = 0





2.3 A Possible Scenario …

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



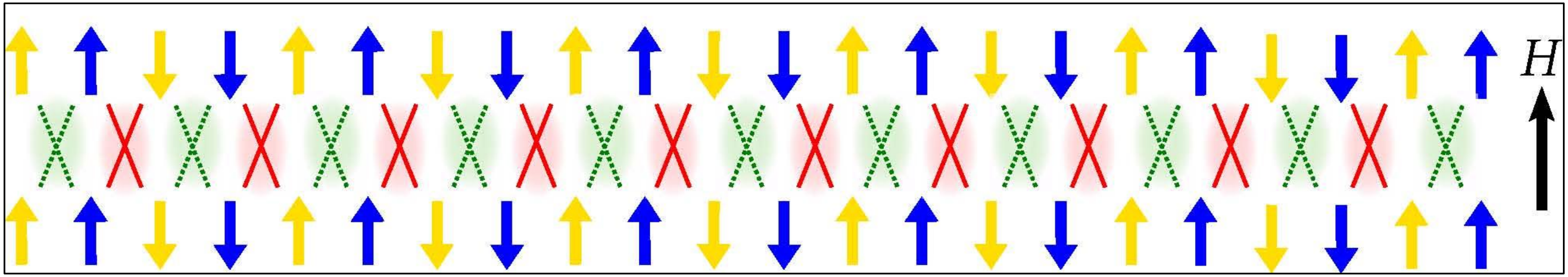
In H=0, long-range dipolar and vector-chiral orders are preserved

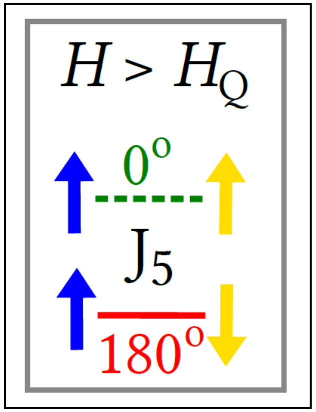
2-soliton bound by intra-chain J1 → vector-chiral order

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

2.3 A Possible Scenario …

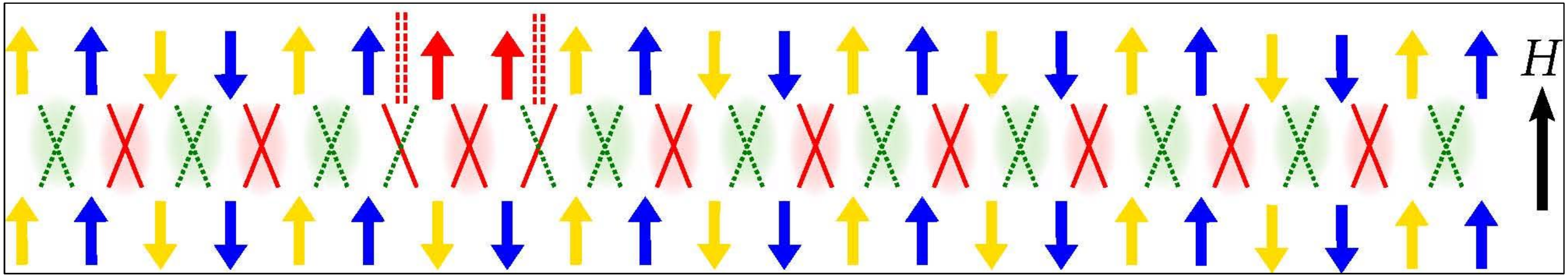
❖ Role of <u>frustrated</u> inter-chain interactions $H > H_Q$



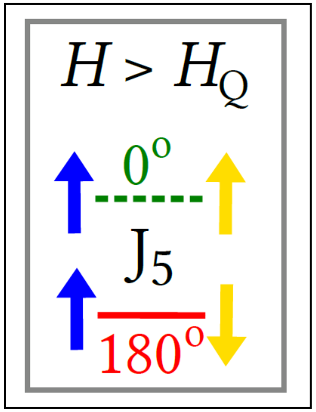


2.3 A Possible Scenario …

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



2-soliton + 2-soliton



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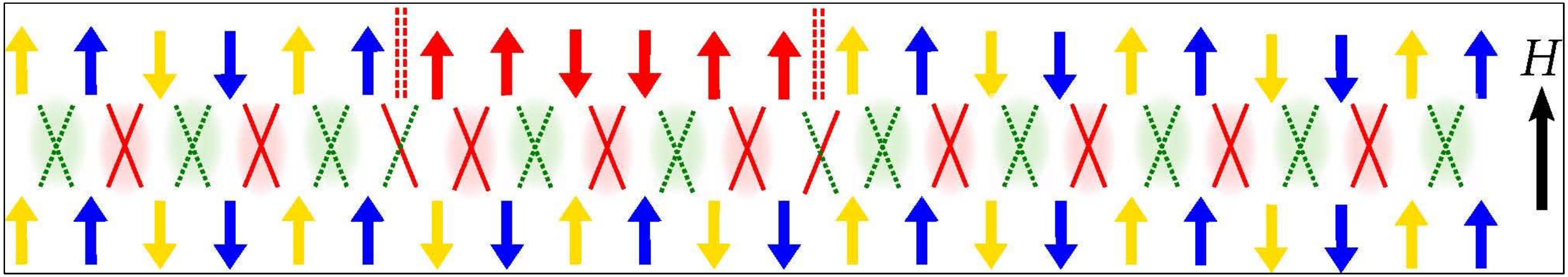
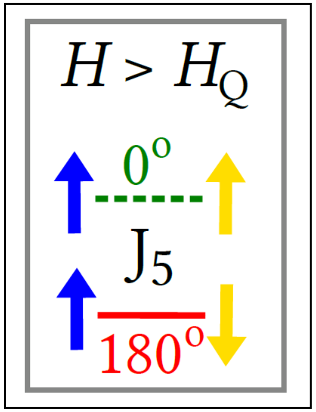
❖ Role of <u>frustrated</u> inter-chain interactions $H > H_Q$





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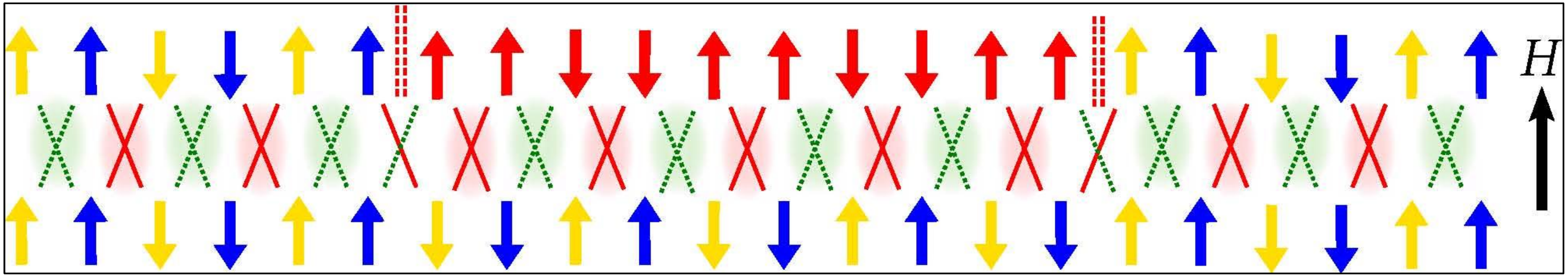
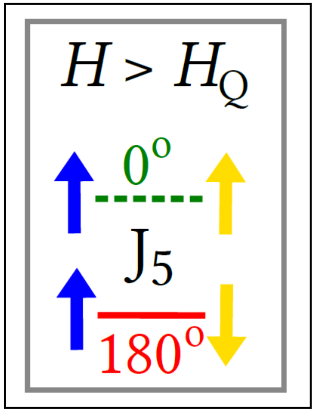
❖ Role of frustrated inter-chain interactions $H > H_Q$





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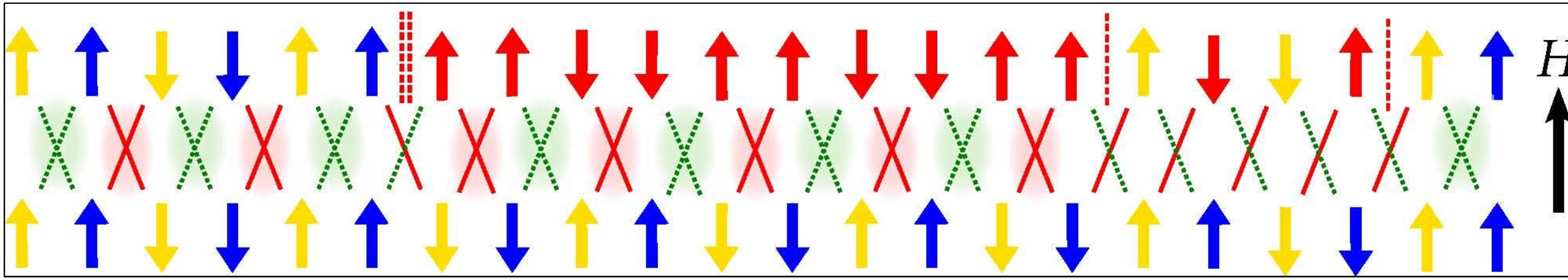
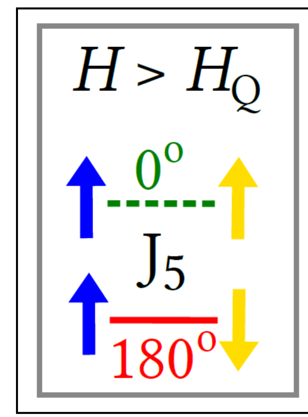
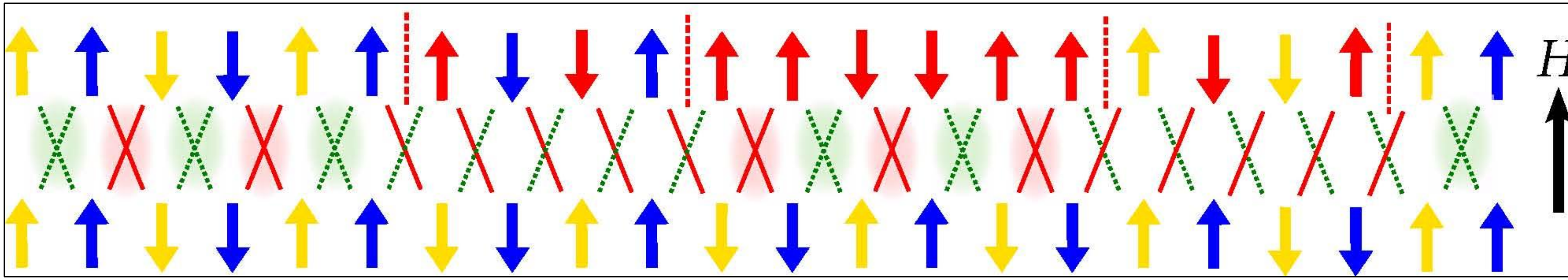
❖ Role of frustrated inter-chain interactions $H > H_Q$

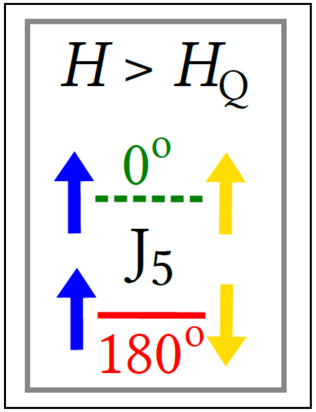




2.3 A Possible Scenario …

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





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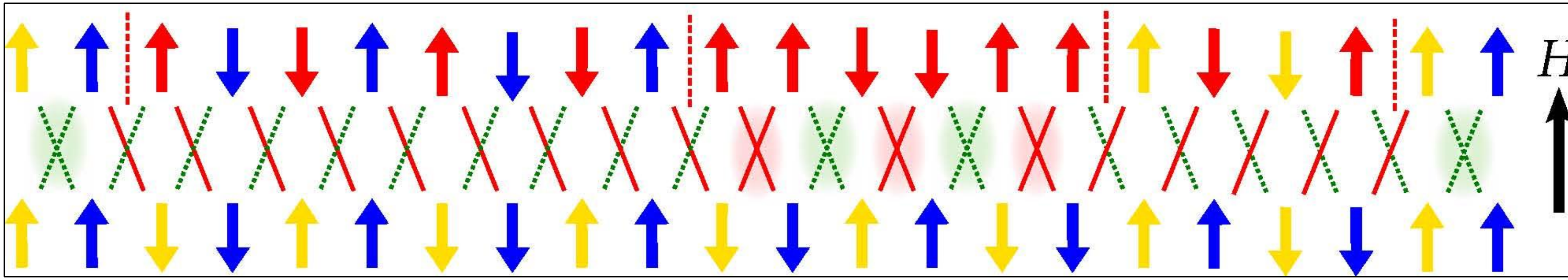
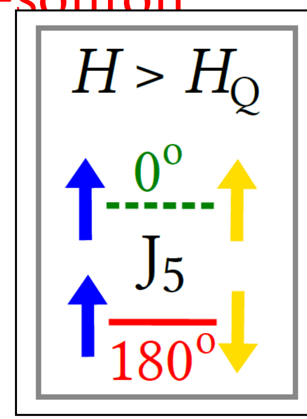
❖ Role of <u>frustrated</u> inter-chain interactions $H > H_Q$

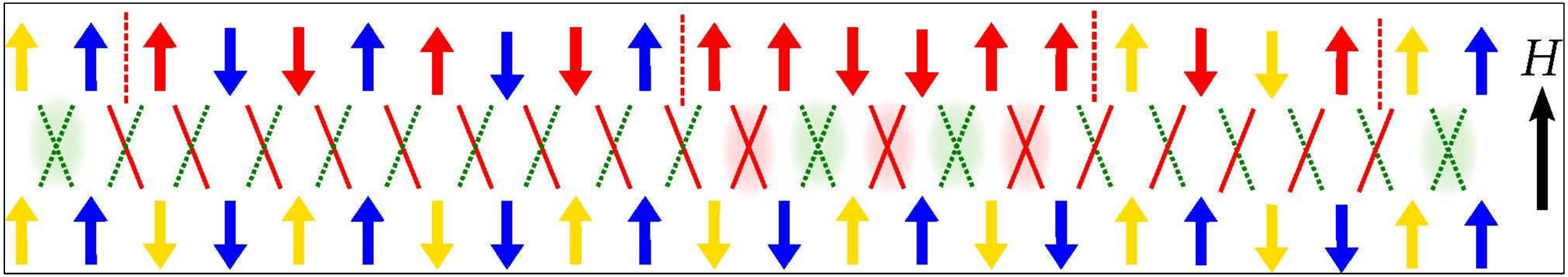


1-soliton 1-soliton 1-soliton 1-soliton



2.3 A Possible Scenario …

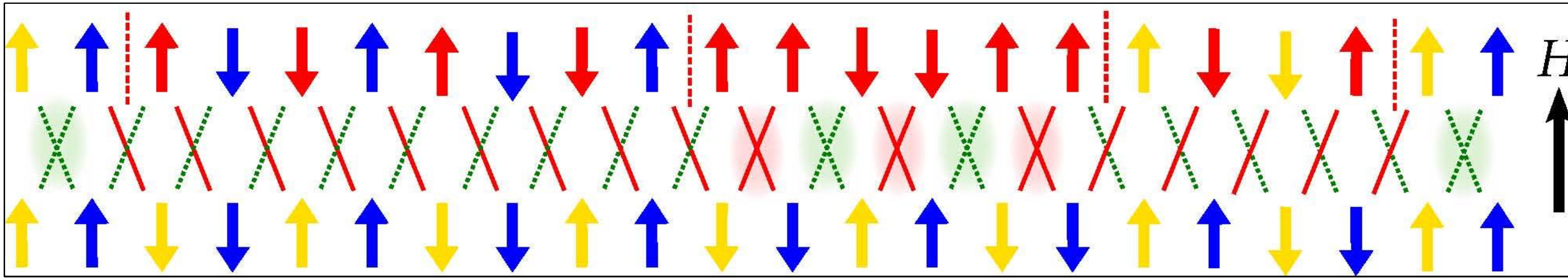
❖ Role of <u>frustrated</u> inter-chain interactions $H > H_Q$



In $H > H_Q$ long-range dipolar order is <u>not favored</u>

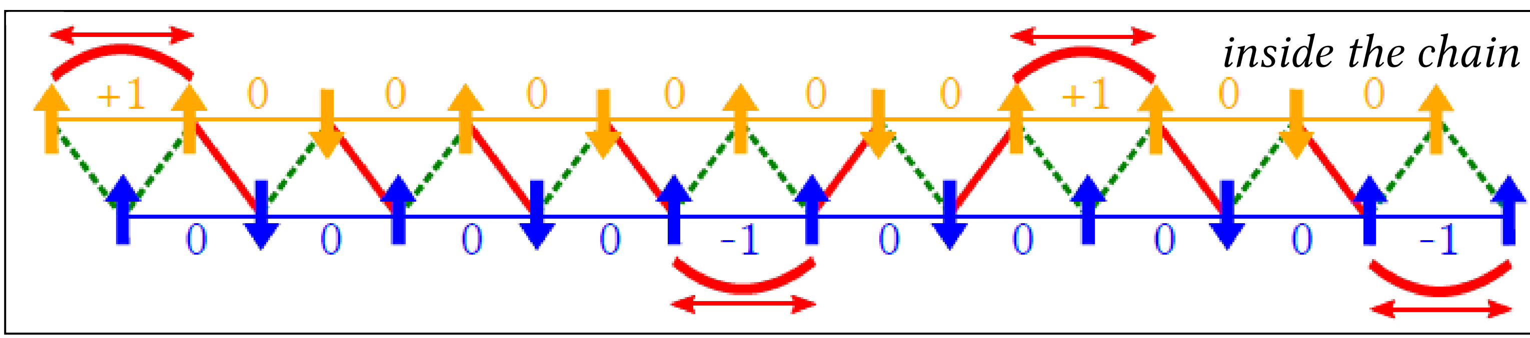
2.3 A Possible Scenario …

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



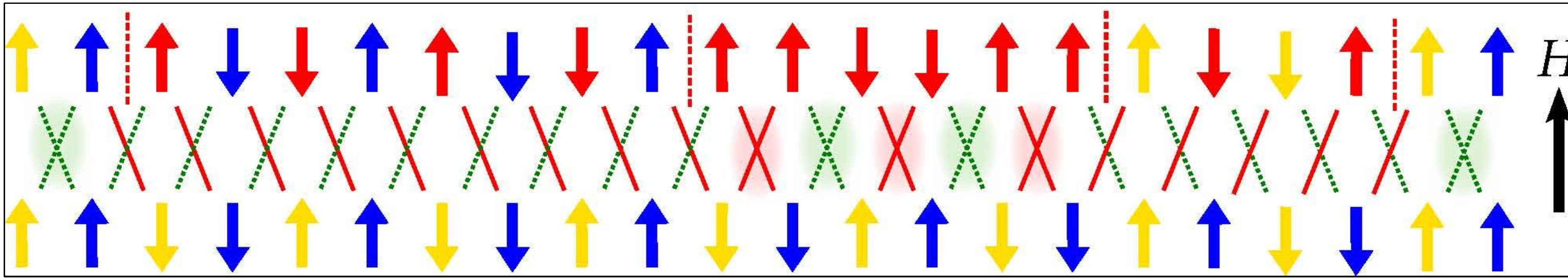
In $H > H_Q$ long-range dipolar order is not favored

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



2.3 A Possible Scenario …

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



In $H > H_Q$ long-range dipolar order is not favored

However, there is a non-local positional order

Conclusions

1. Role of frustrated inter-chain interactions

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