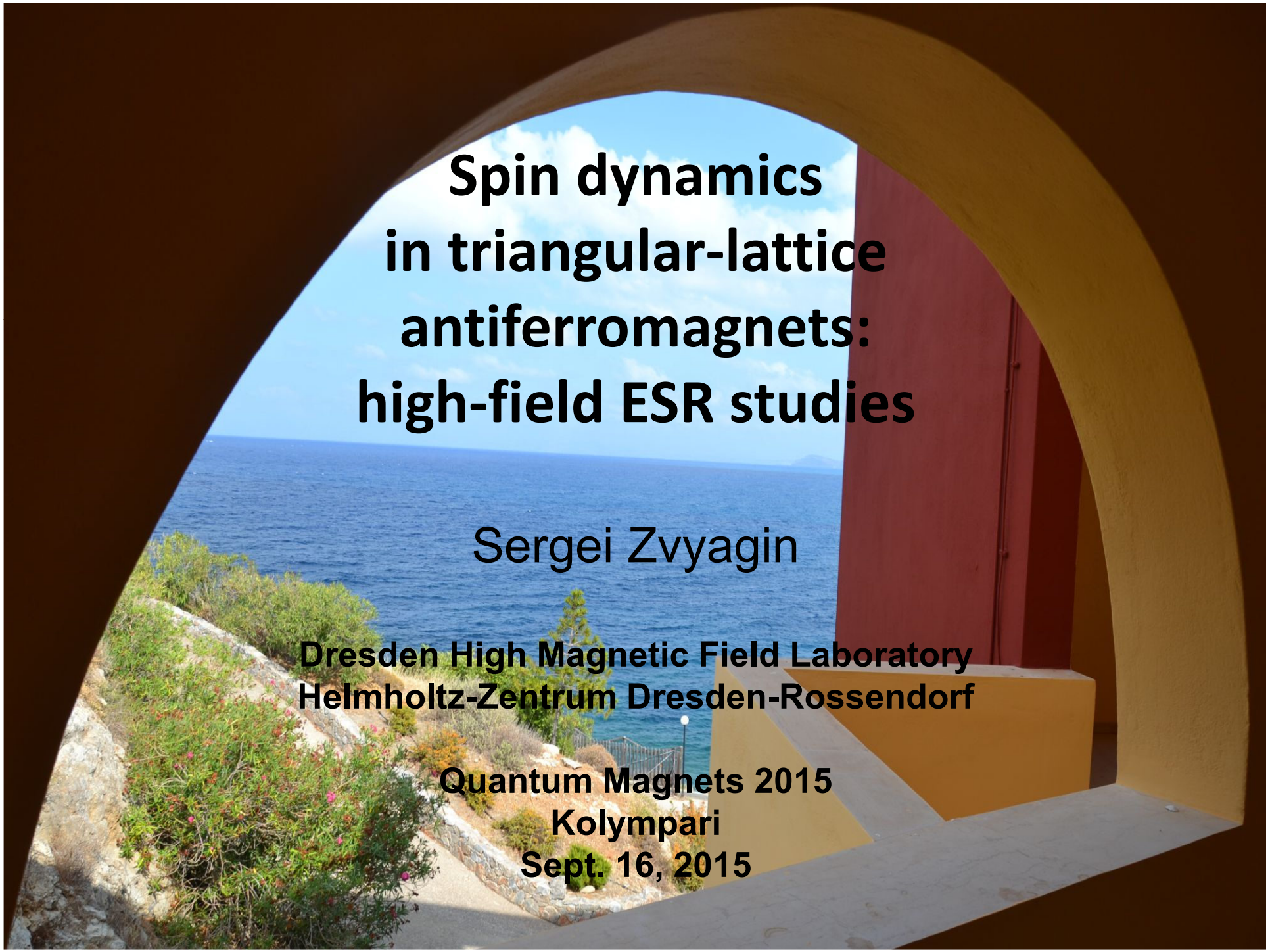

Spin dynamics in triangular-lattice antiferromagnets: high-field ESR studies

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Dresden High Magnetic Field Laboratory
Helmholtz-Zentrum Dresden-Rossendorf

Quantum Magnets 2015
Kolympari
Sept. 16, 2015

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T. Fujita
M. Hagiwara **[pulsed-field ESR measurements of Cs_2CuBr_4]**

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A. Smirnov P.L. Kapitza Institute for Physical Problems, Moscow, Russia
T. Soldatov **[collaboration on Cs_2CuCl_4]**

D. Kamenskyi HLD-HZDR, Dresden, Germany

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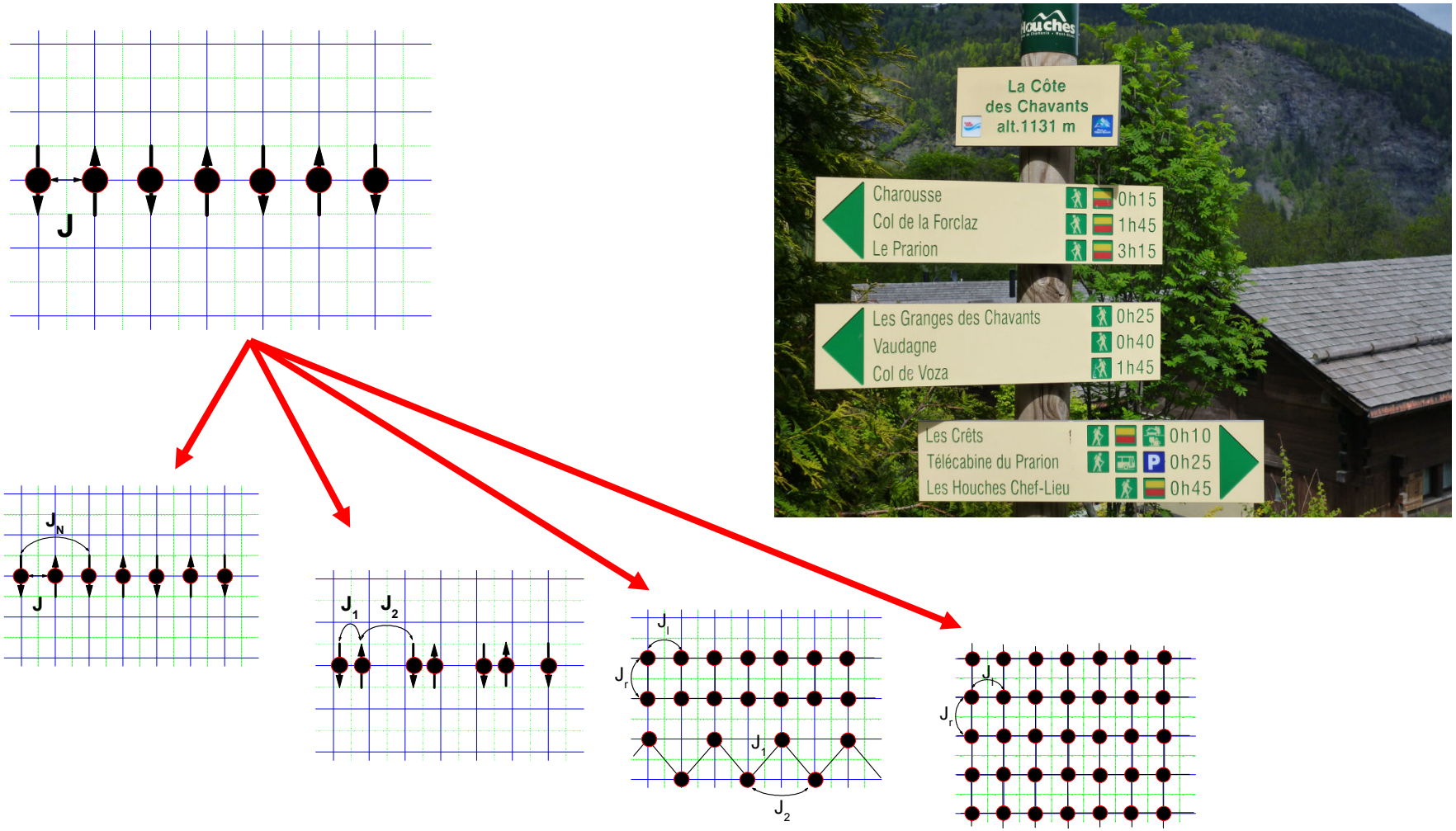
R. Hu Brookhaven Natl. lab, USA
H. Ryu
C. Petrovic **[synthesis of Cs_2CuBr_4 samples]**

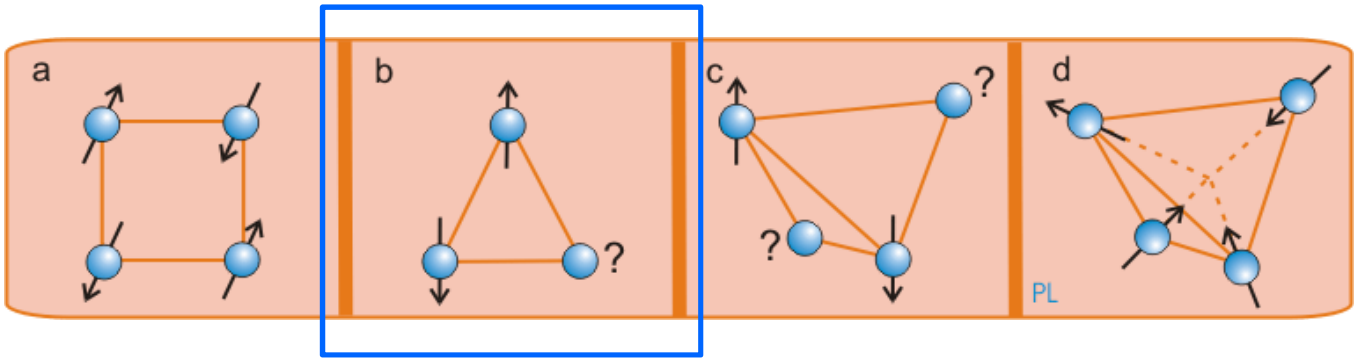
M. Zhitomirsky CEA-INAC/UJF, Grenoble, France **[theory]**

Diversity of low-D magnets

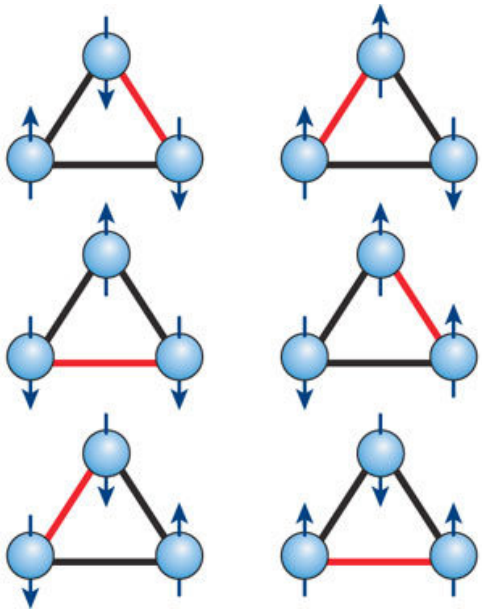


- A unique way to design quantum magnets
- Ideal playground for testing various theoretical concepts

Geometrical frustrations: a new way to look at magnetic interactions



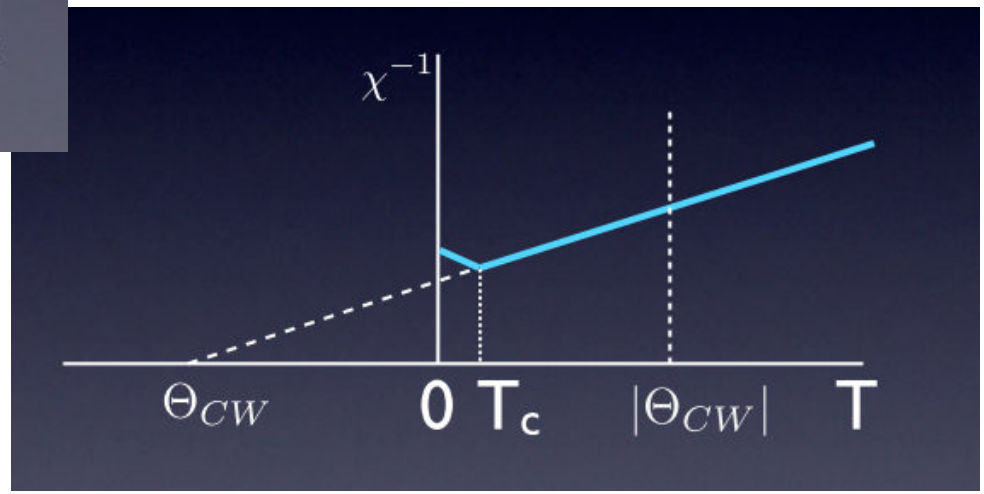
What is special:



- Degeneracy of the ground state
- Instead of ordering, the system fluctuates amongst the many degenerate states, even at $T \rightarrow 0$
- Fine tuning of the degeneracy – unusual magnetic properties

Empirical signature of frustration

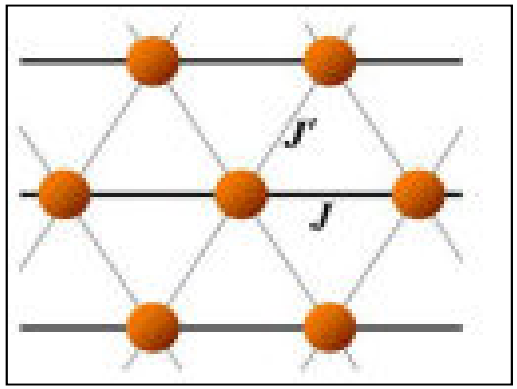
$$f = \frac{|\Theta_{CW}|}{T_c}$$

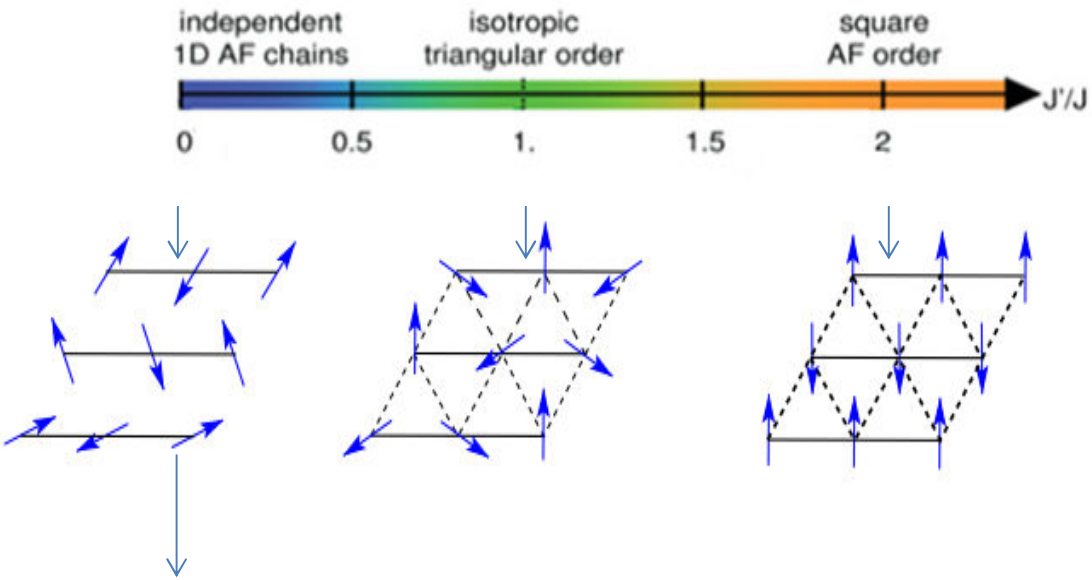


Open questions

1. Nature of the ground state in the disordered state
2. Elementary excitations in the ordered/disordered phase
3. Competition/cooperation between magnetic order, quantum fluctuations and the geometrical frustration
4. Realization of exotic phases: spin liquid, spin ice, magnetic monopoles, etc.
5. Rich phase diagram; very attractive for high-field research

Triangular-lattice AFs: properties of the ground state





1D collinear phase is predicted to exist up to *J'/J* ~ 0.2 – 0.8

Starykh et al., PRB 82, 014421(2010)
Heidarian et al., PRB 80, 012404 (2009)
Weng ey al., PRB 74, 012407 (2006)

Question on the critical *J'/J* ratio.

Some examples of frustrated magnets (Ramirez, 1994)

Compound	Magnetic lattice	$-\theta_{cw}$ (K)	T_c (K)	f	Ordered state	Electronic configuration
wo-dimensional magnets						
VCl_2	triangular	437	36	12	AF	$3d^3$
$NaTiO_2$	triangular	1000	<2	>500	—	$3d^1$
$LiCrO_2$	triangular	490	15	33	AF	$3d^3$
$Gd_{0.8}La_{0.2}CuO_2$	triangular	12.5	0.7	16	SG	$4f^7$
$SrCr_8Ga_4O_{19}$	kagome	515	3.5	150	SG	$3d^3$
$KCr_3(OH)_6(SO_4)_2$	kagome	70	1.8	39	AF	$3d^3$
hree-dimensional magnets						
$ZnCr_2O_4$	B-spinel	390	16	24	AF	$3d^3$
K_2IrCl_6	FCC	321	3.1	10	AF	$5d^5$
FeF_3	B-spinel	240	15	16	AF	$3d^5$
$CsNiFeF_5$	B-spinel	210	4.4	48	SG	$3d^8$, $3d^5$
$MnIn_2Te_4$	zinc blende	100	4	25	SG	$3d^5$
$Gd_3Ga_5O_{12}$	garnet	2.3	<0.03	>100	—	$4f^7$
Sr_2NbFeO_6	perovskite	840	28	30	SG	$3d^4$
Ba_2NbVO_6	perovskite	450	15	30	SG	$3d^3$

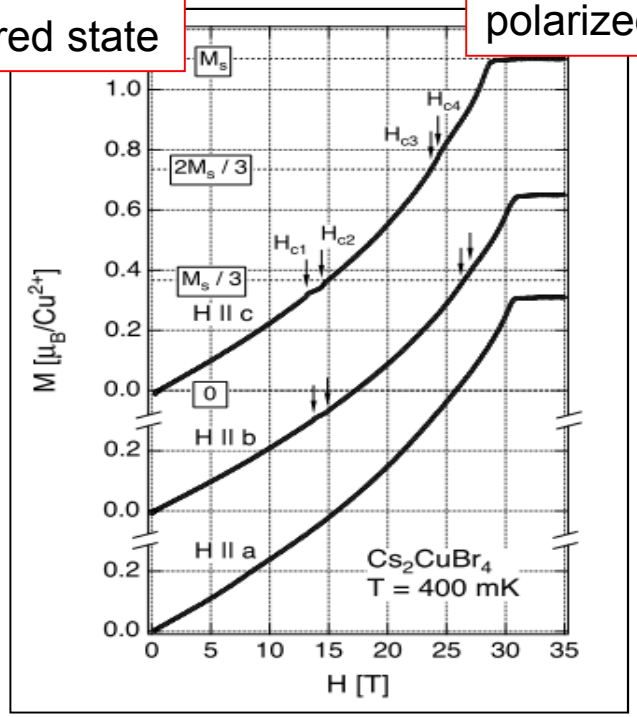
Cs_2CuBr_4 and Cs_2CuCl_4 (J ~ 10 K) is the most suitable materials for research

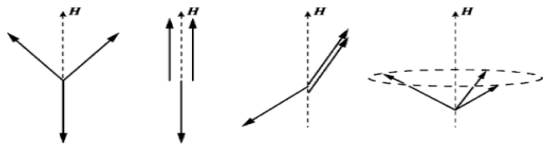
Cs_2CuBr_4: magnetic properties

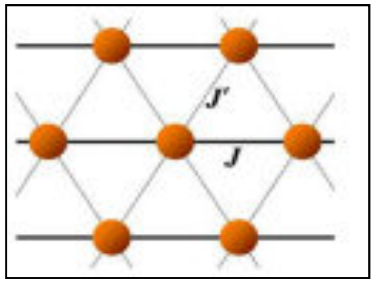
Ono et al., JPSJ 74, 135 (2005)

Incommensurate (helix) ordered state

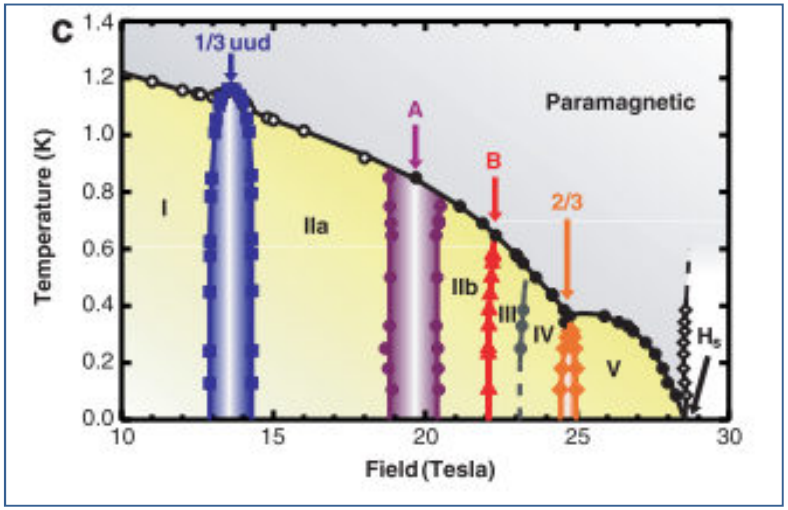
Fully spin polarized state







Fortune et al., PRL 102, 257201 (2009)

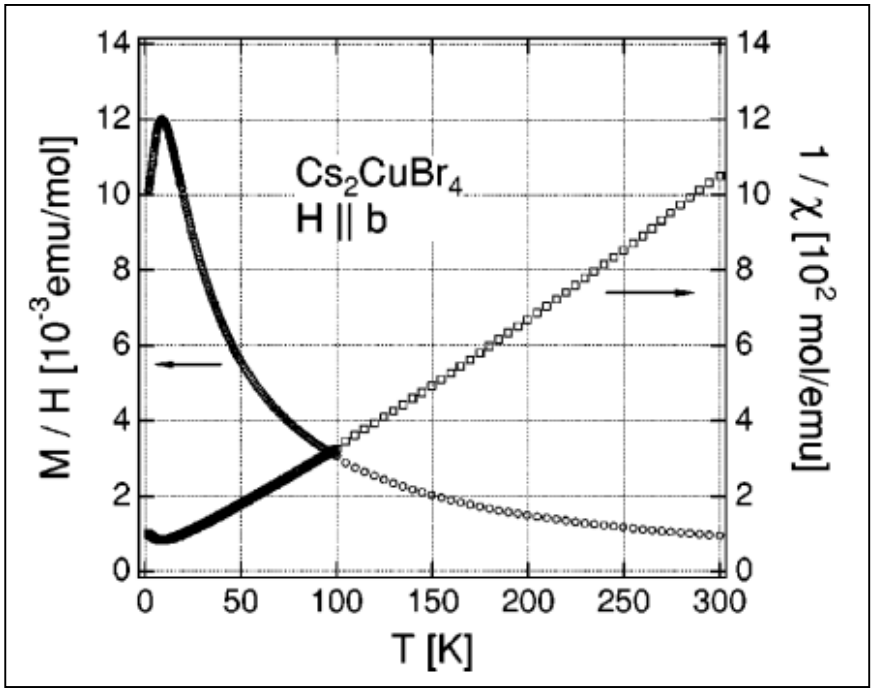


To describe the phase diagram quantitatively the information on the exchange parameters is extremely important!

Uncertainty in exchange parameters:

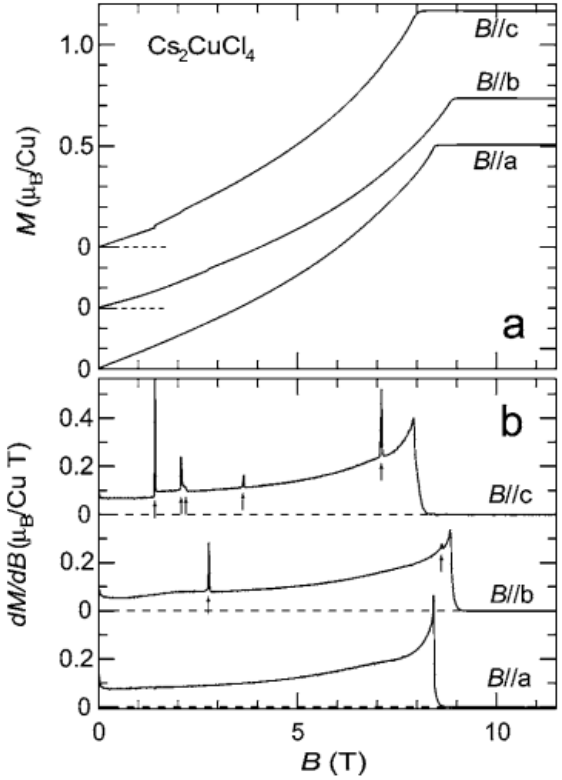
$J \sim 10 - 30$ K; $J'/J \sim 0.4 - 0.8$

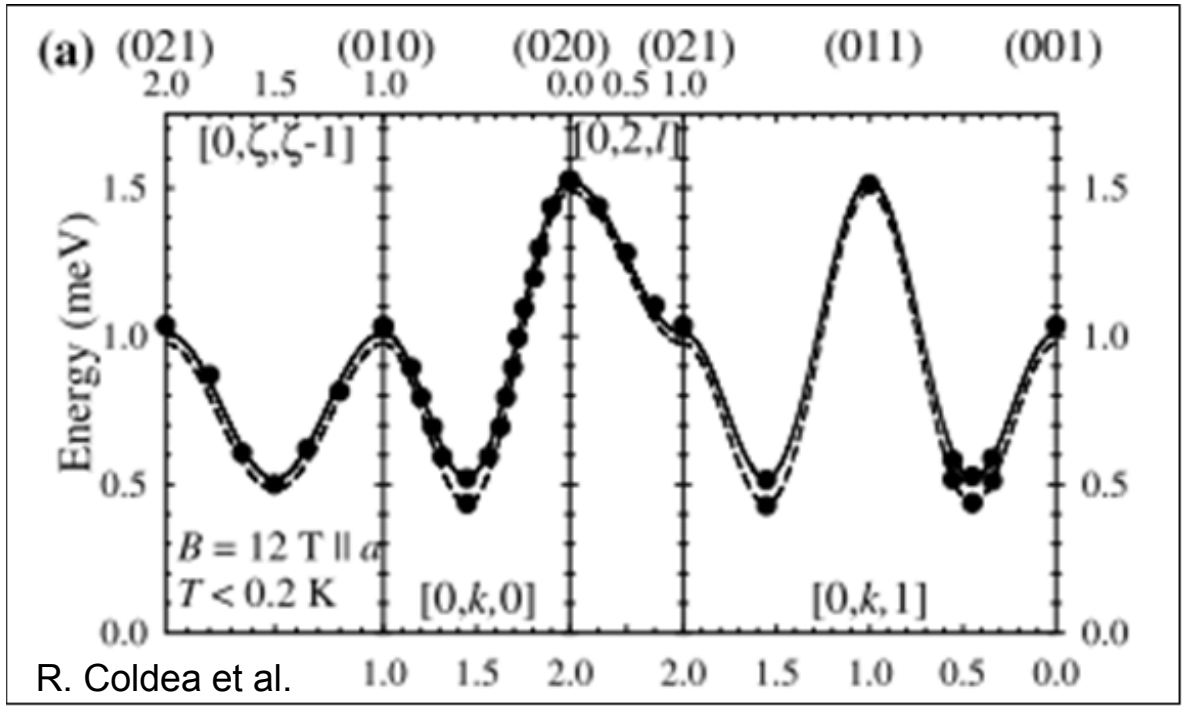
1.Ono et al., J. Phys. Soc. Jpn. 74, 135 (2005)
2. Foyevtsova et al., PRB 83, 125126 (2011)
3.Ono et al., J. Phys.: Cond. Matt. 16, S773 (2004) ; Weihong et al., PRB 59, 14367 (1999)



Effect of quantum fluctuations makes quantitate description extremely challenging.

Solution: to go to very high magnetic fields ($H > H_{sat}$), where the quantum fluctuations are fully suppressed and the classical spin-wave theory can be used





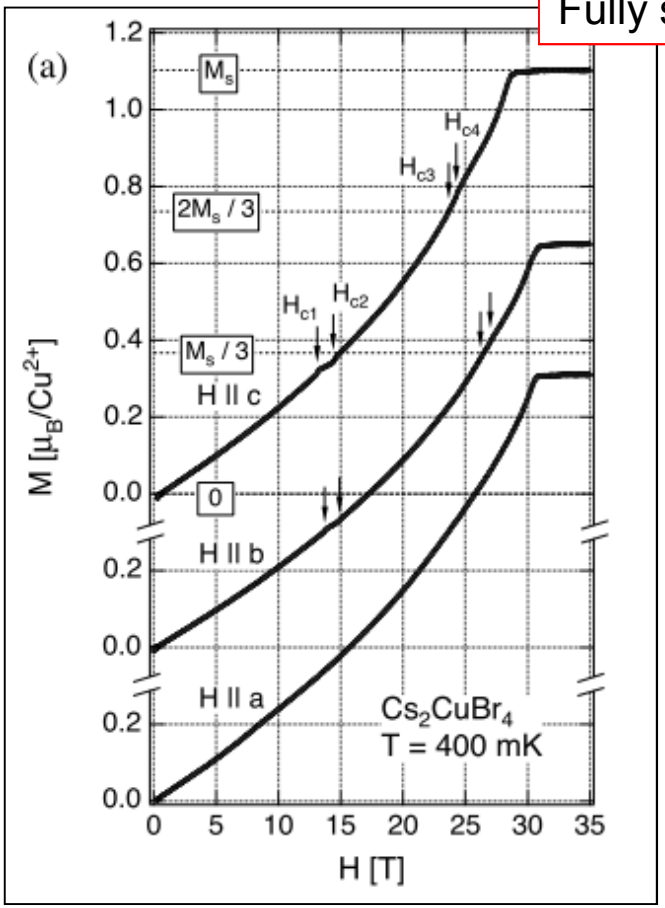
R. Coldea et al.

J = 4.3 K; J'/J = 0.34

Isostructural Cs_2CuCl_4: inelastic neutron scattering in the fully spin polarized state

Magnetization in Cs_2CuBr_4

Fully spin polarized state

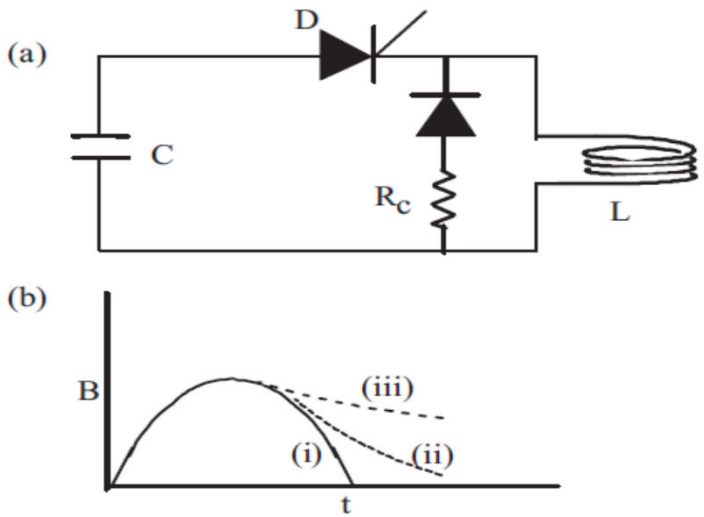


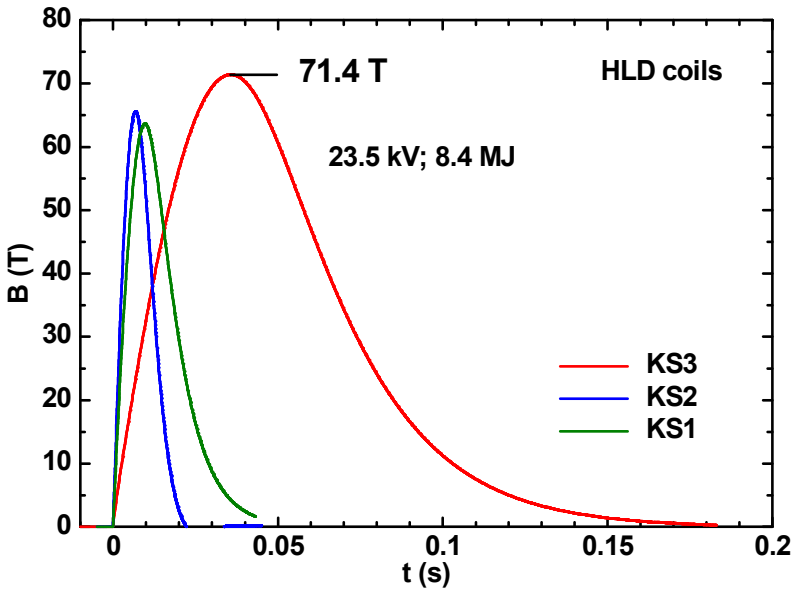
H_{sat} ~ 30 T

ESR is a powerful tool to probe spin dynamics in such a high filed.

Ono et al., JPSJ 74, 135 (2005)

Dresden High Magnetic Field Laboratory

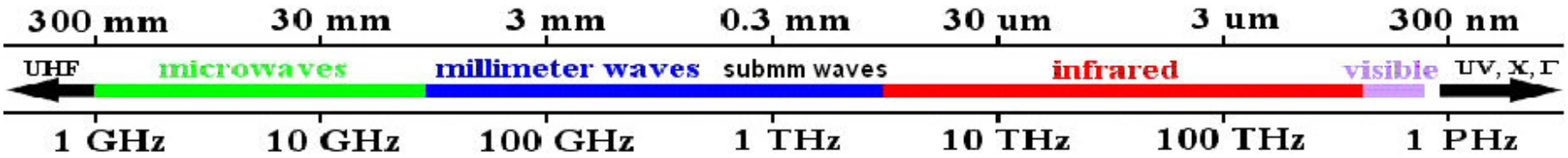


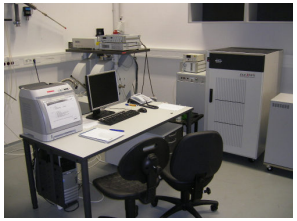


THz GAP: ~0.3 - 3 THz

Microwaves

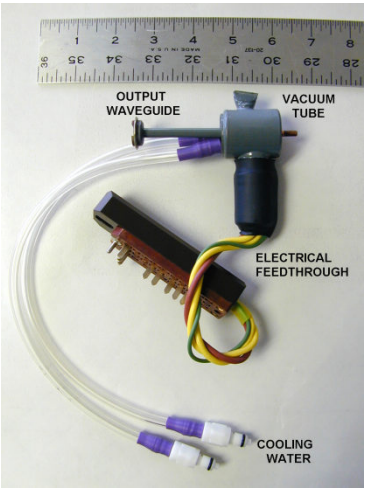
Optics



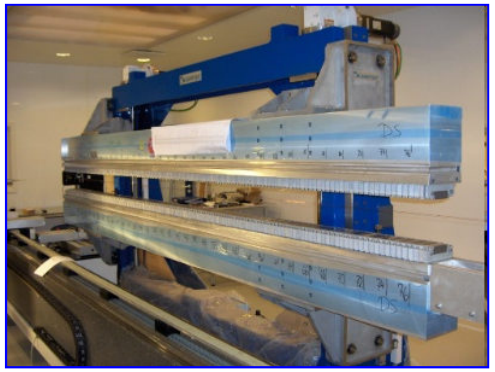


X-band Bruker spectrometer

Gunn/VDI Diodes



BWO

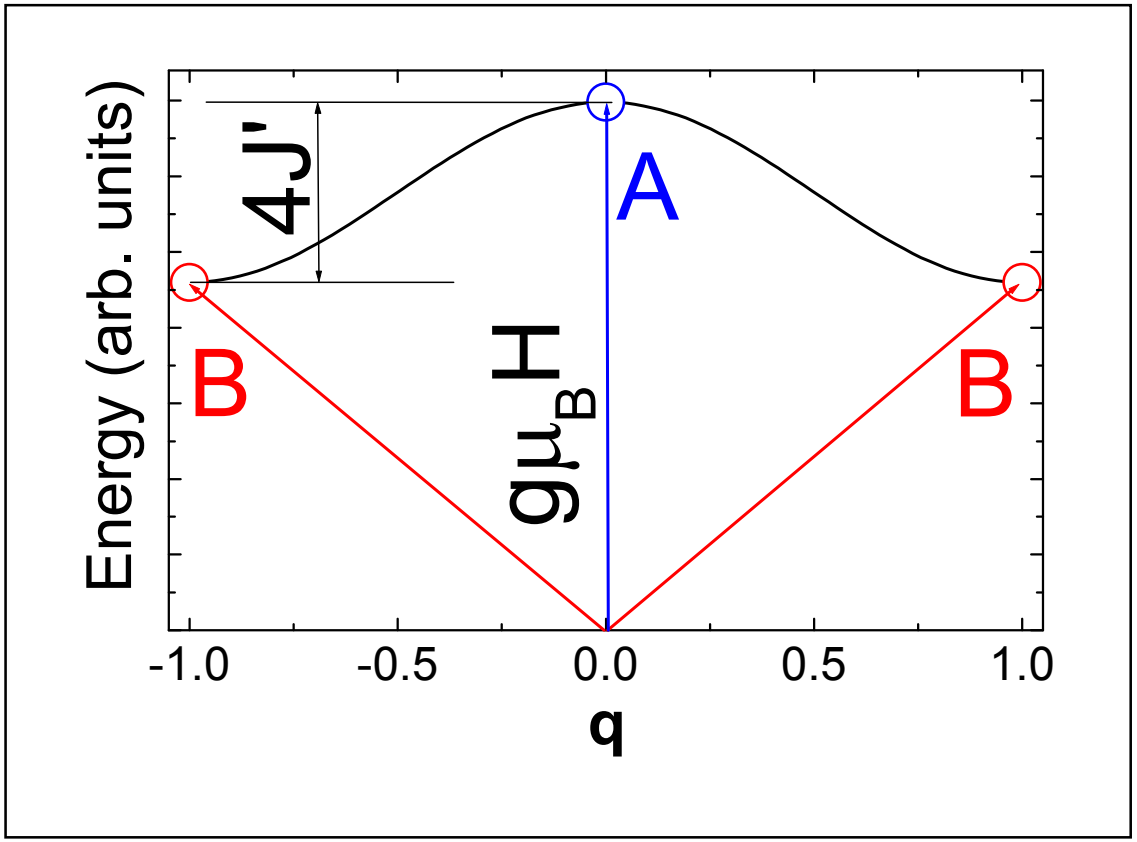


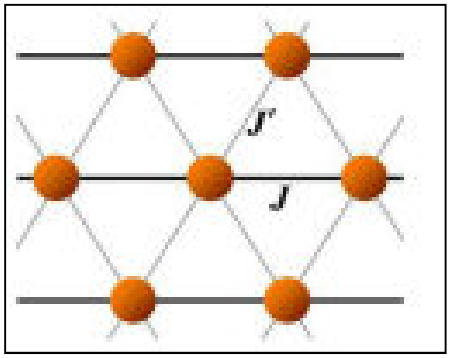
Free Electron Laser

9 GHz, 50 GHz – 1.3 THz (conventional), 1.2 – 16 THz (FEL)

Fields: 16 T (DC superconducting) 70 T (pulsed);

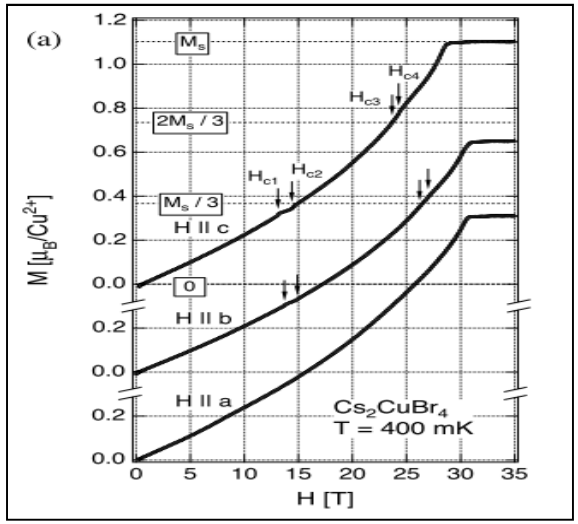
Excitation spectrum in the spin-polarized phase



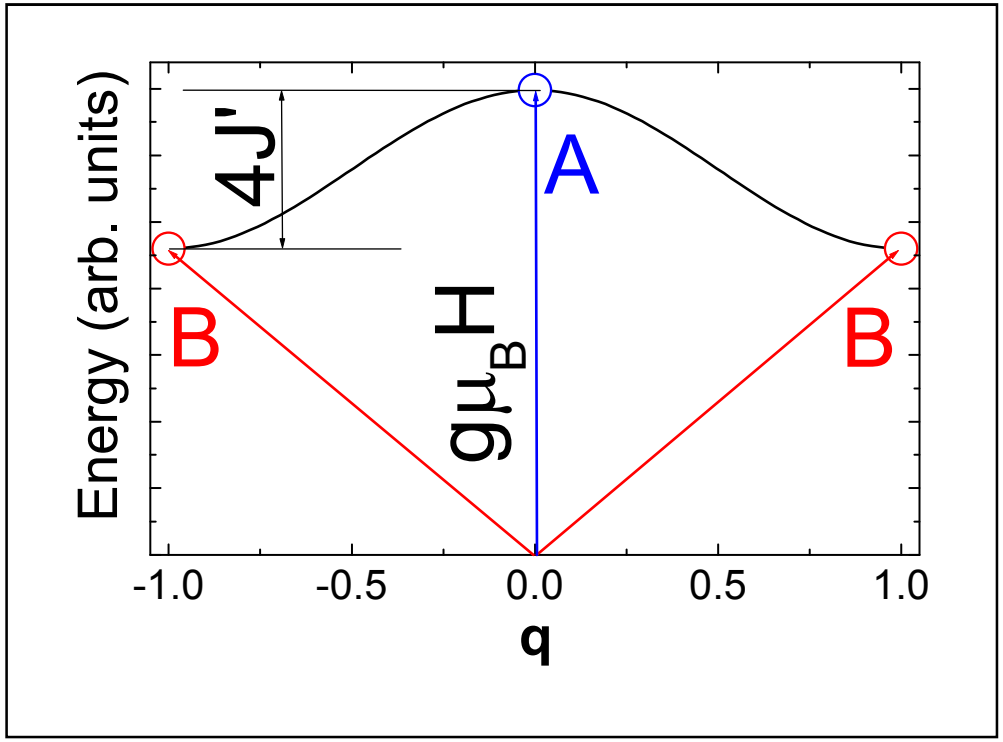


Observation of mode B is possible due to staggered DM interaction

Magnetic excitations in a triangular-lattice AF in the fully spin polarized phase: spin-wave theory approach

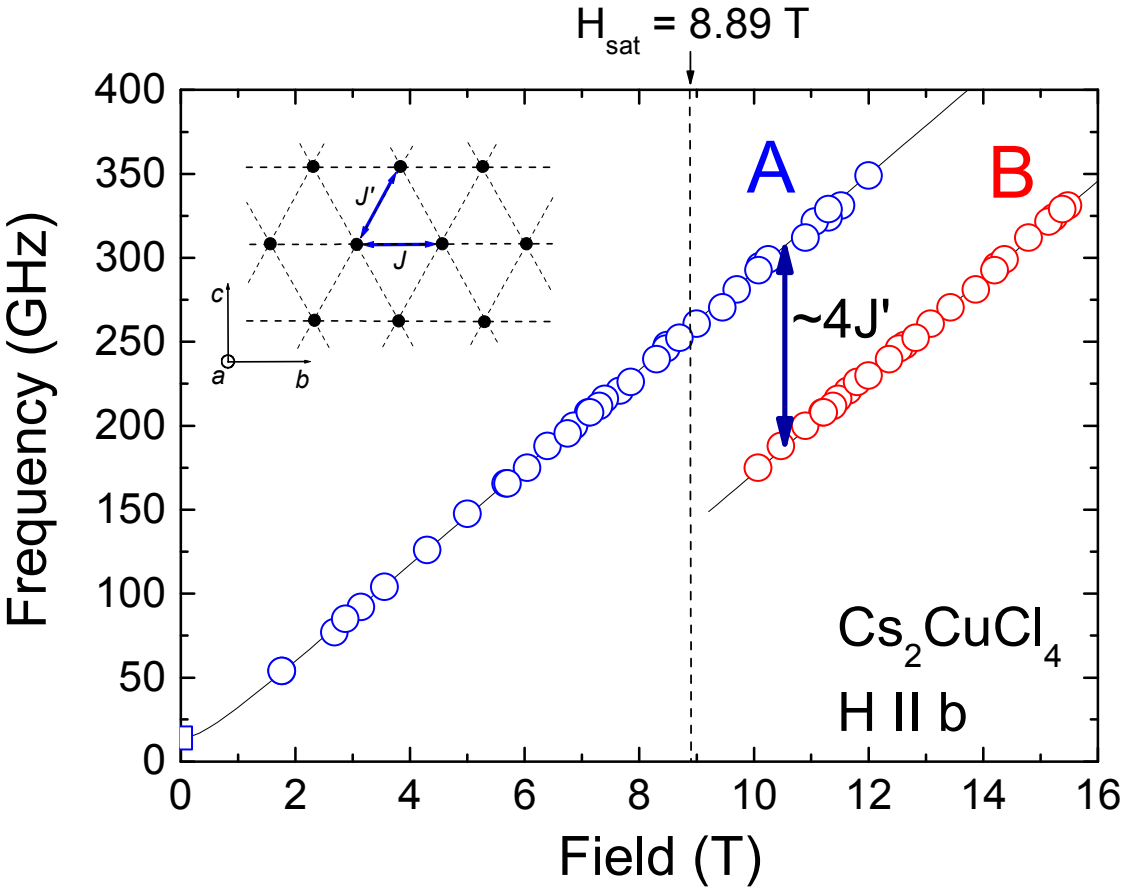


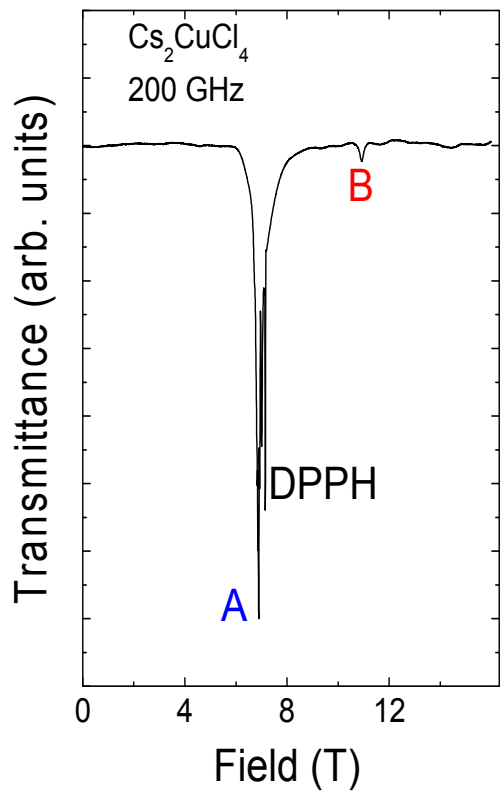
$$h\Delta\nu = 4J'$$
$$g\mu_B H_{sat} = 2J(1+J'/2J)^2$$



Staggered DM interaction leads to the effective zone folding and allows transitions at q = π. Elementary excitations are magnons.

ESR in the isostructural Cs_2CuCl_4 in the fully spin polarized phase



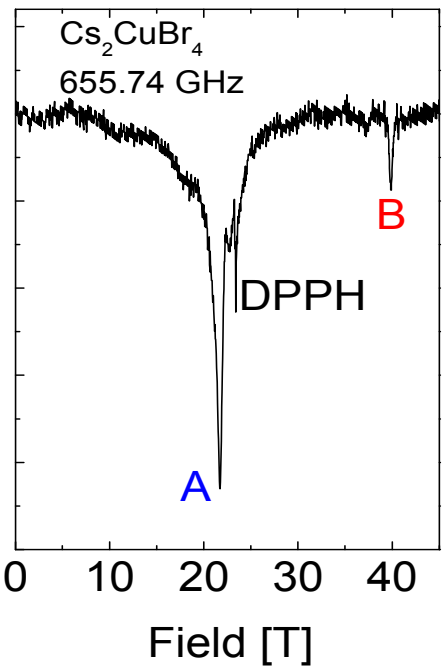


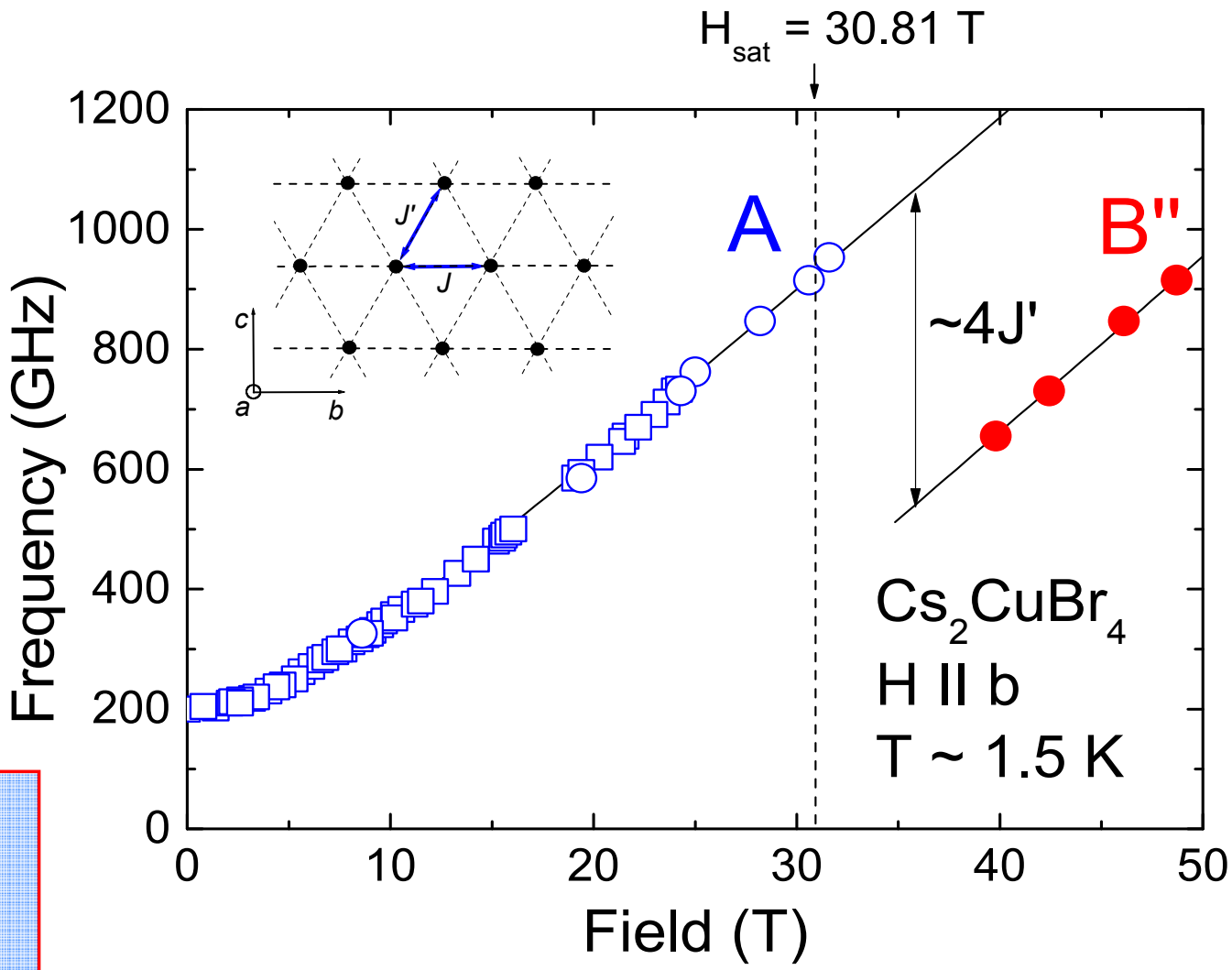
Test for validity of the used approach – very good agreement:

ESR: J = 4.7 K; J'/J = 0.30(3)

INS: J = 4.3 K; J'/J = 0.34(2)

First direct determination of the exchange parameters in Cs_2CuBr_4



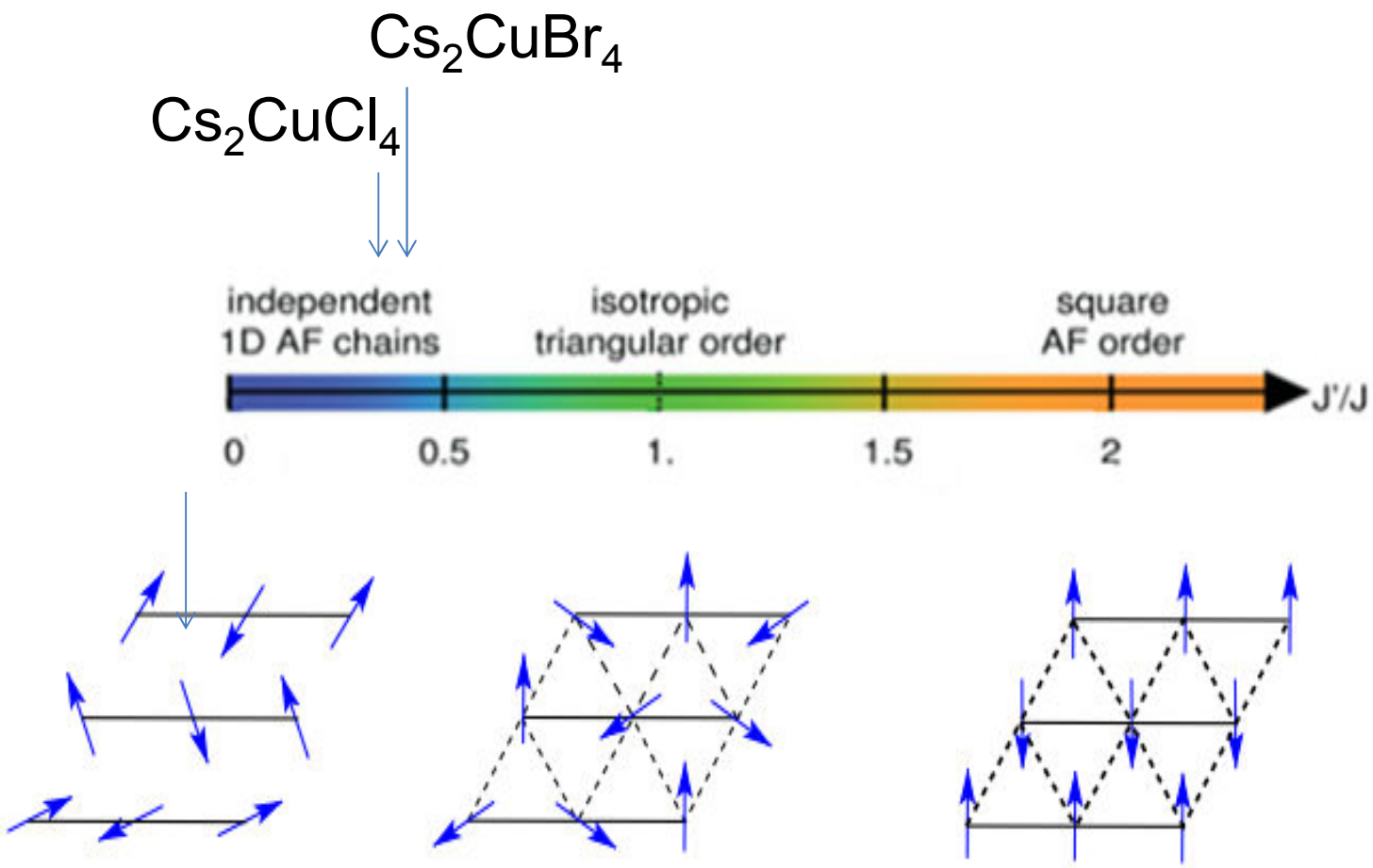


$J/k_B = 14.9(7)$ K

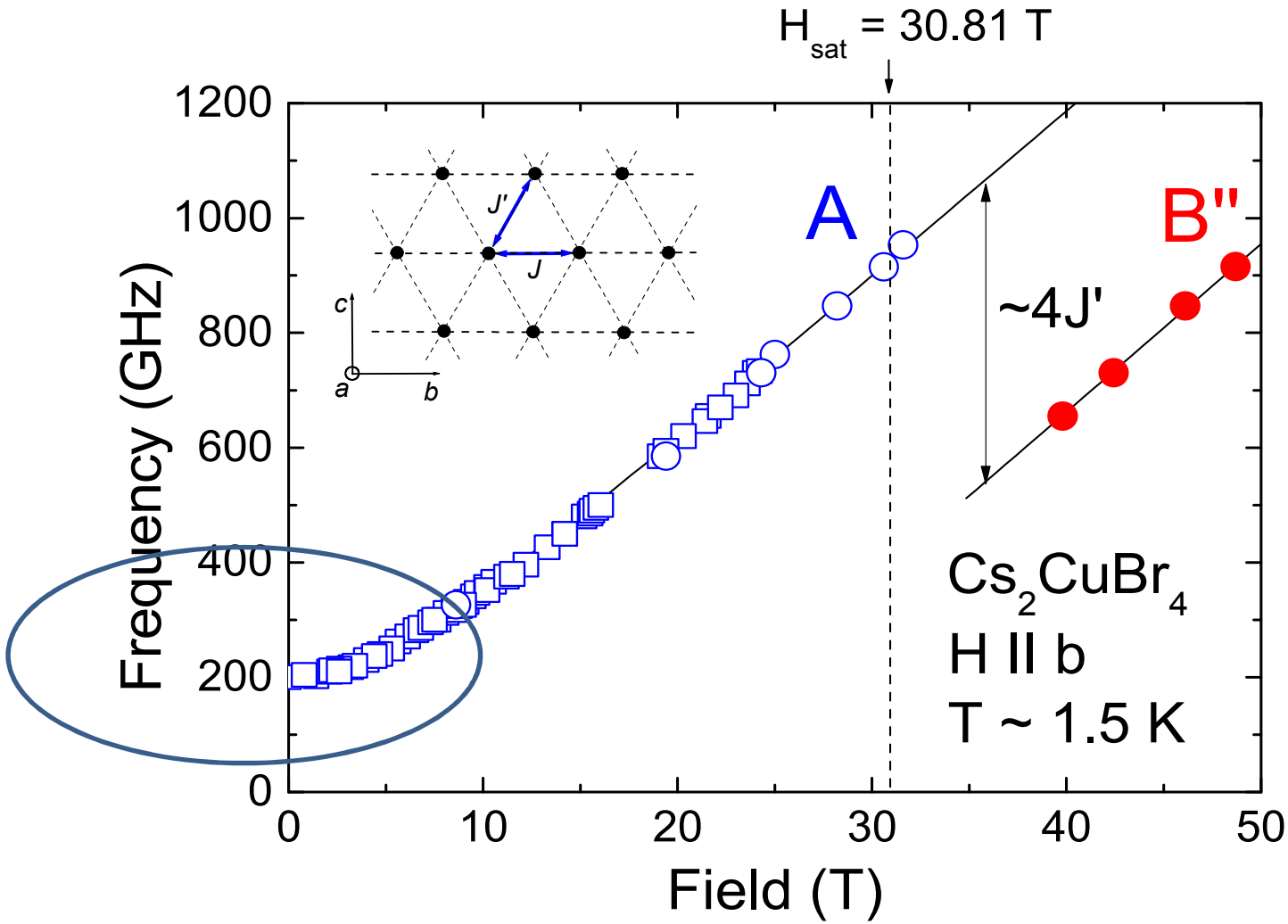
$J'/k_B = 6.1(3)$ K

$J'/J = 0.41$

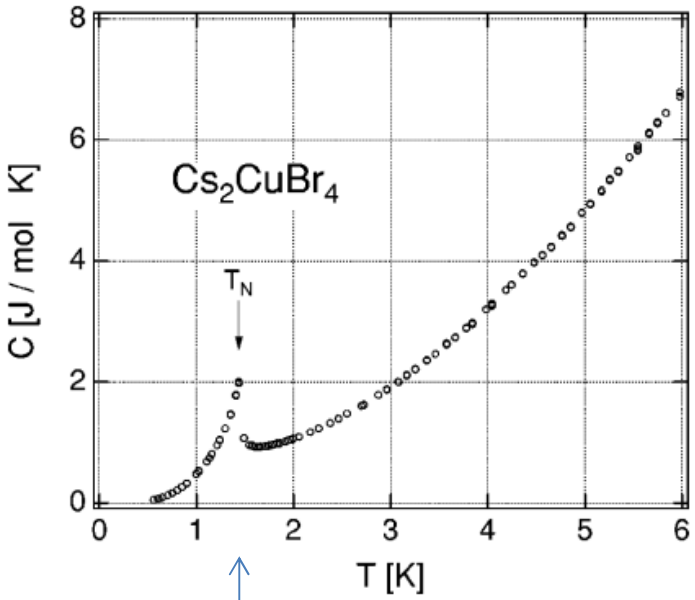
Triangular-lattice AFs: properties of the ground state



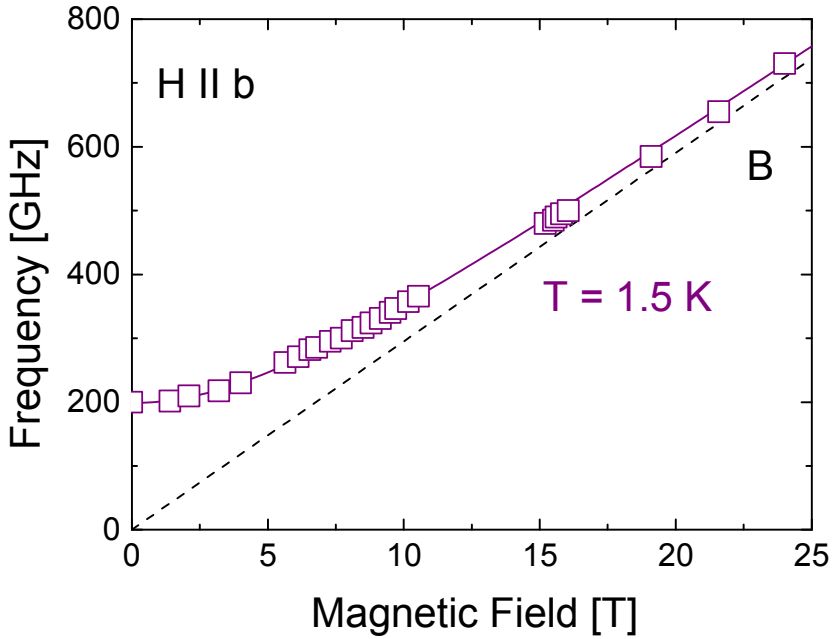
What about the origin of the gap?



Excitations in Cs_2CuBr_4 in the AF-ordered state

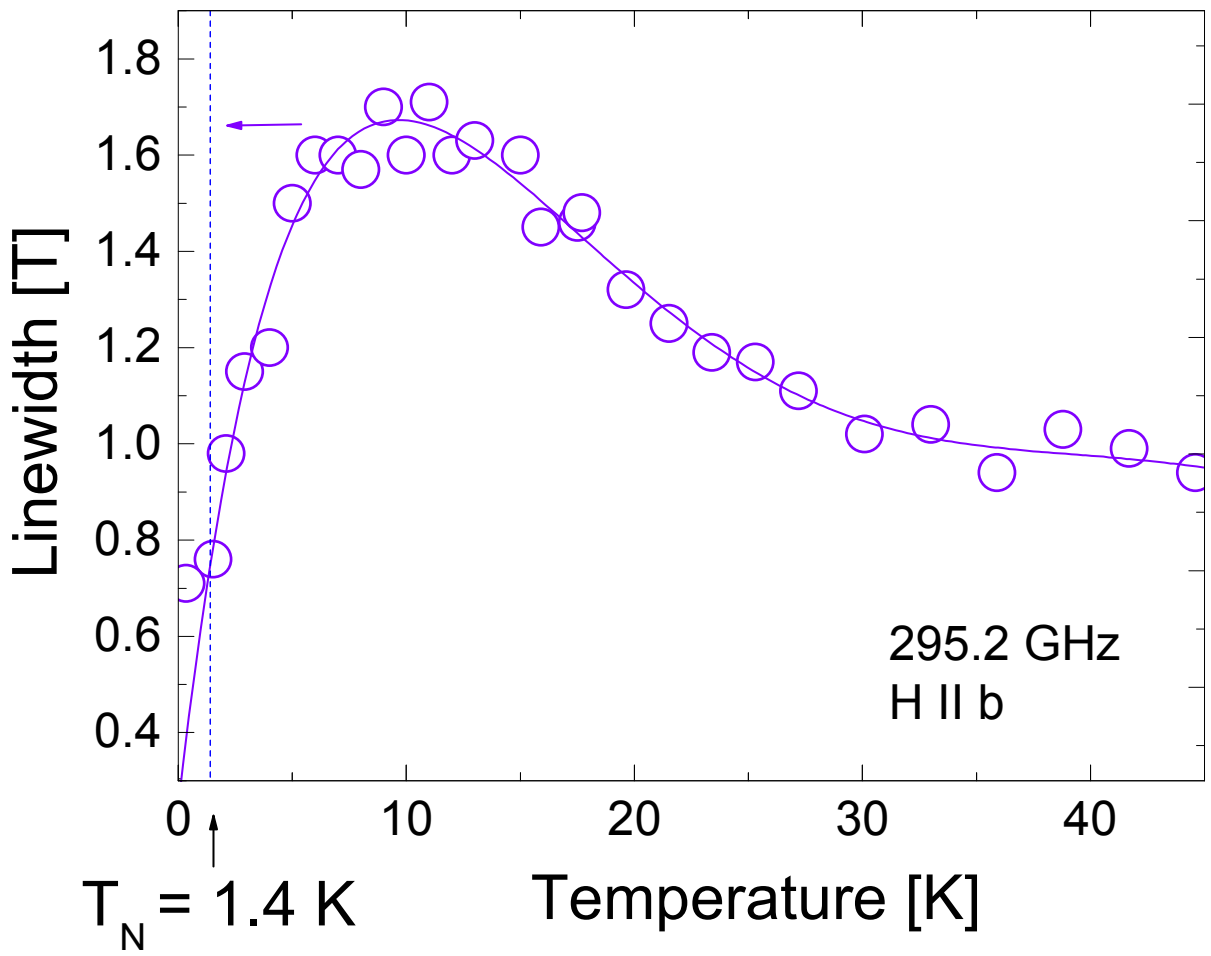


T_N = 1.3 K

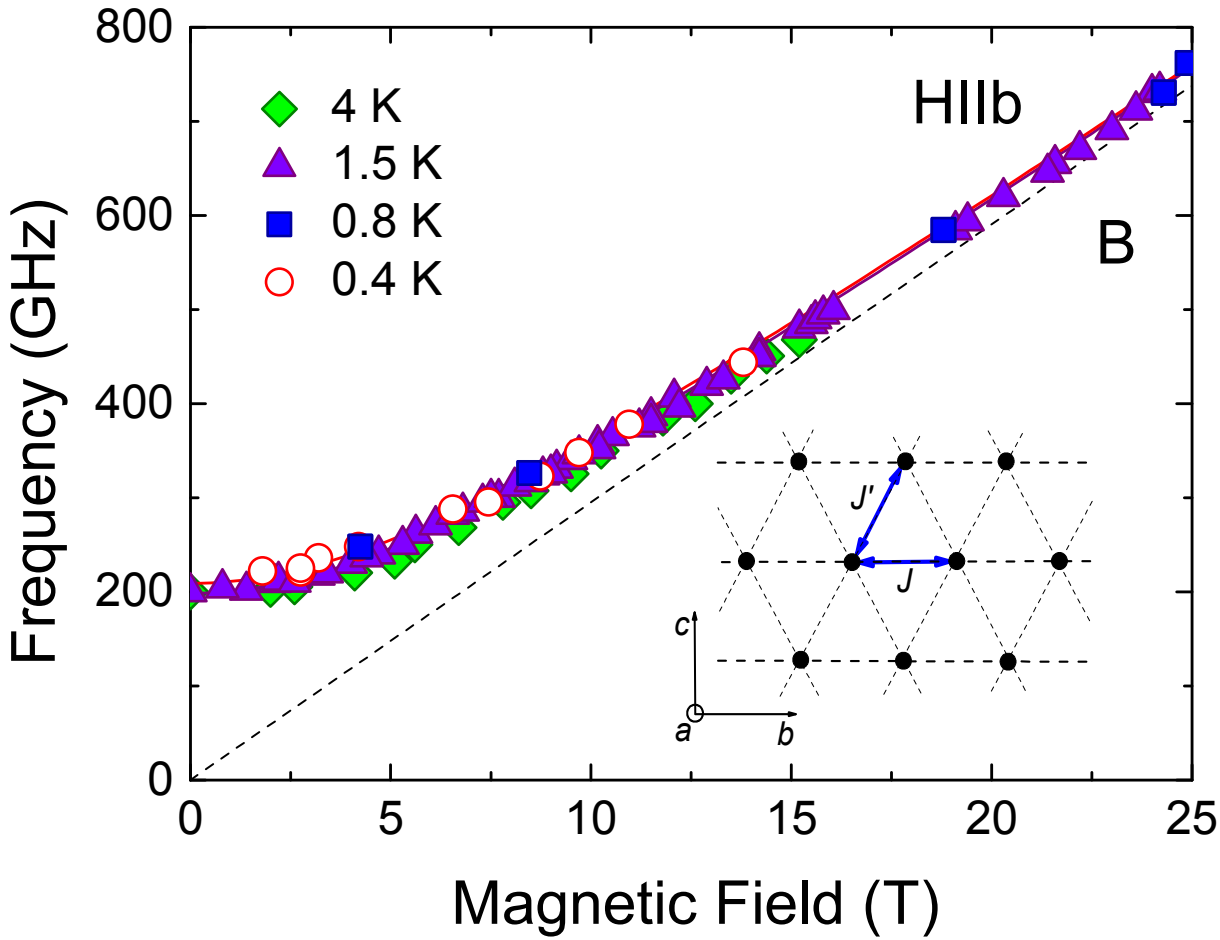


What happens with the excitation spectrum at the transition to AF-ordered state?

Absence of a ESR line-width anomaly at T_N.c

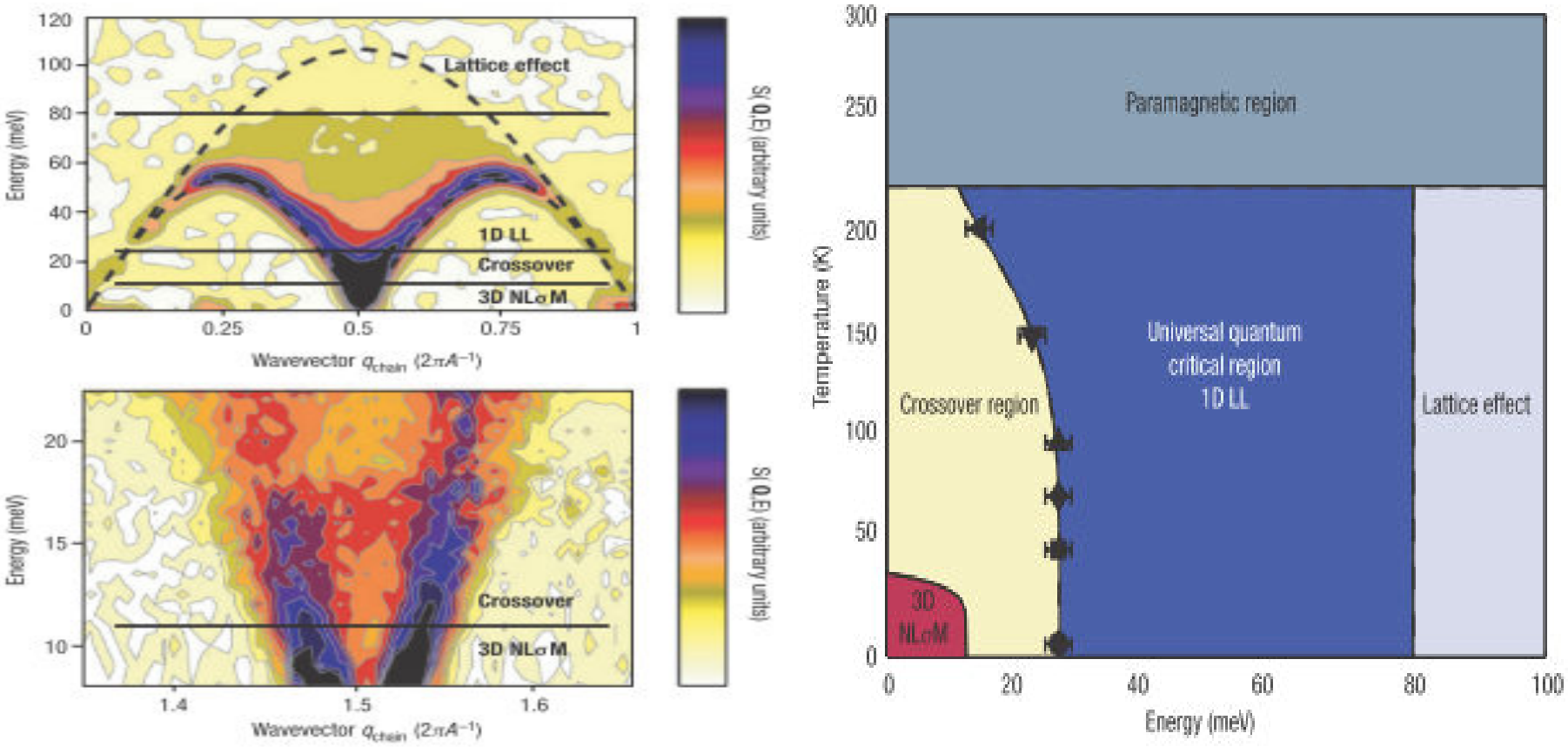


Excitations in Cs_2CuBr_4 above and below T_N



No significant changes while going o magnetically ordered state

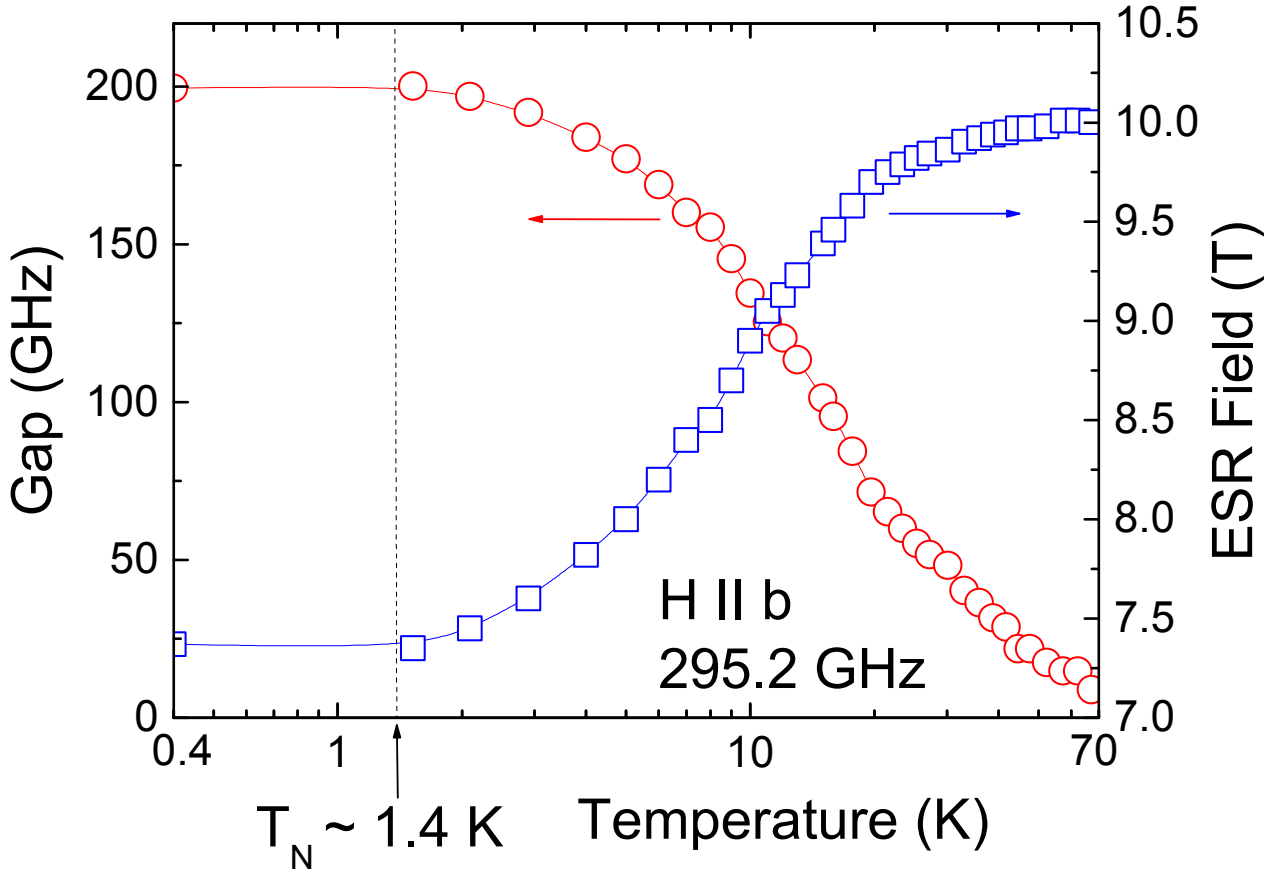
Magnetic excitations in the quasi-1D spin-liquid material $KCuF_3$



Lake et al., Nat. Mat. 4, 329 (2005)

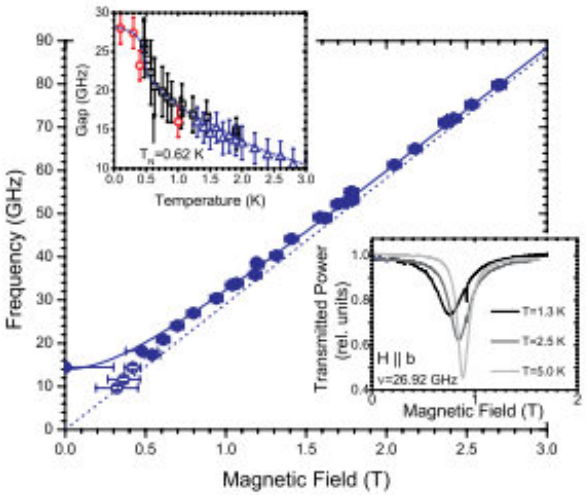
Other examples: Cs_2CuCl_4 [Coldea et al. PRB 68, 134424 (2003)]; $CsNiCl_3$ [Zaliznyak et al., PRL 87, 017202 (2011)]

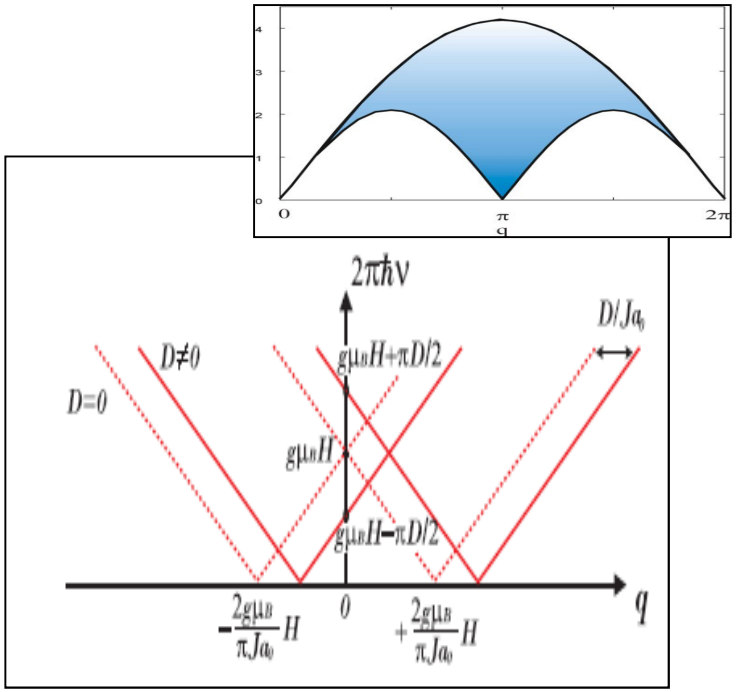
The gap is present above as well as below T_N

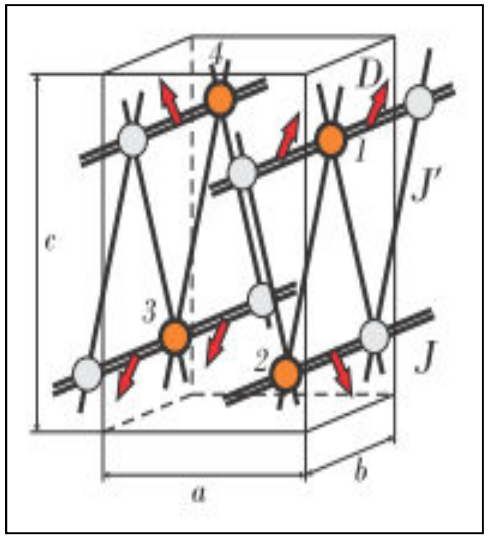


The gap is a property of the short-range-order correlated state

The origin of the gap in the isostructural Cs_2CuCl_4







Spin chains and uniform Dzyaloshinskii-Moriya interaction

Gap = 14 GHz = 0.7 K ~ DM interaction

Povarov, Smirnov et al., PRL 107, 037204 (2011)

Cs_2CuCl_4:
Spin chains and the uniform Dzyaloshinskii-Moriya interaction

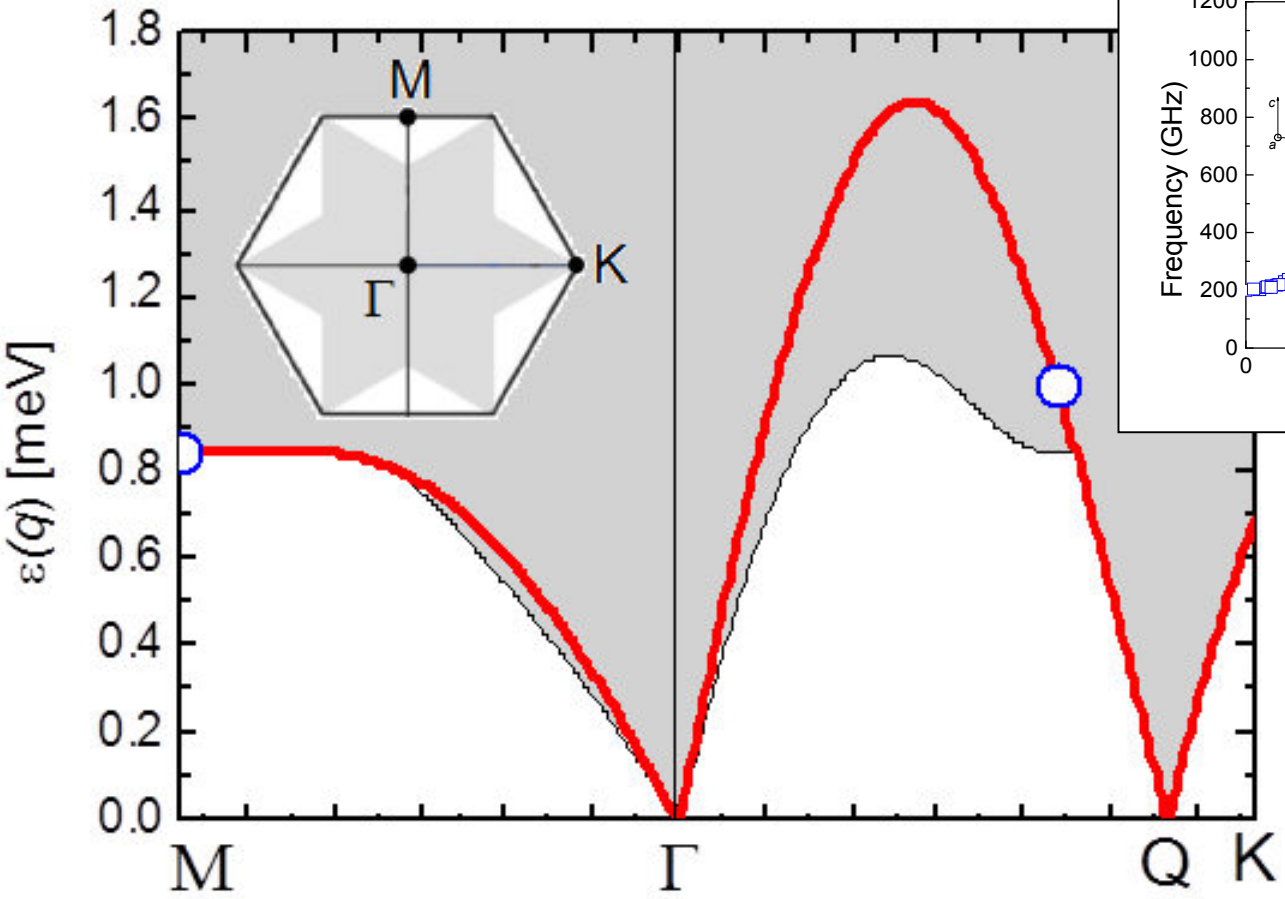
Gap = 14 GHz = 0.7 K ~ 0.1 J ~ DM interaction

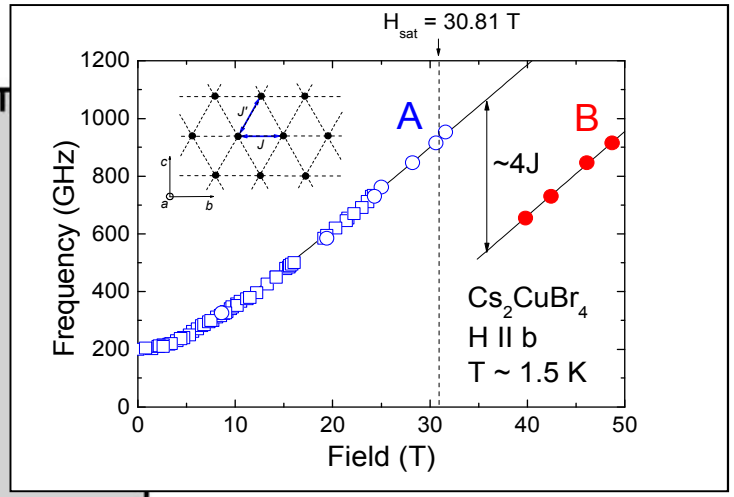
Cs_2CuBr_4:

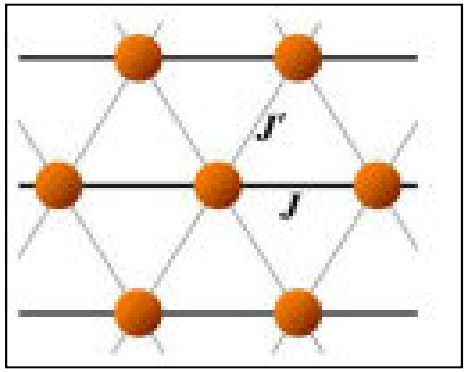
Gap = 200 GHz = 9.6 K ~ 0.6 J

What is the origin of the gap?

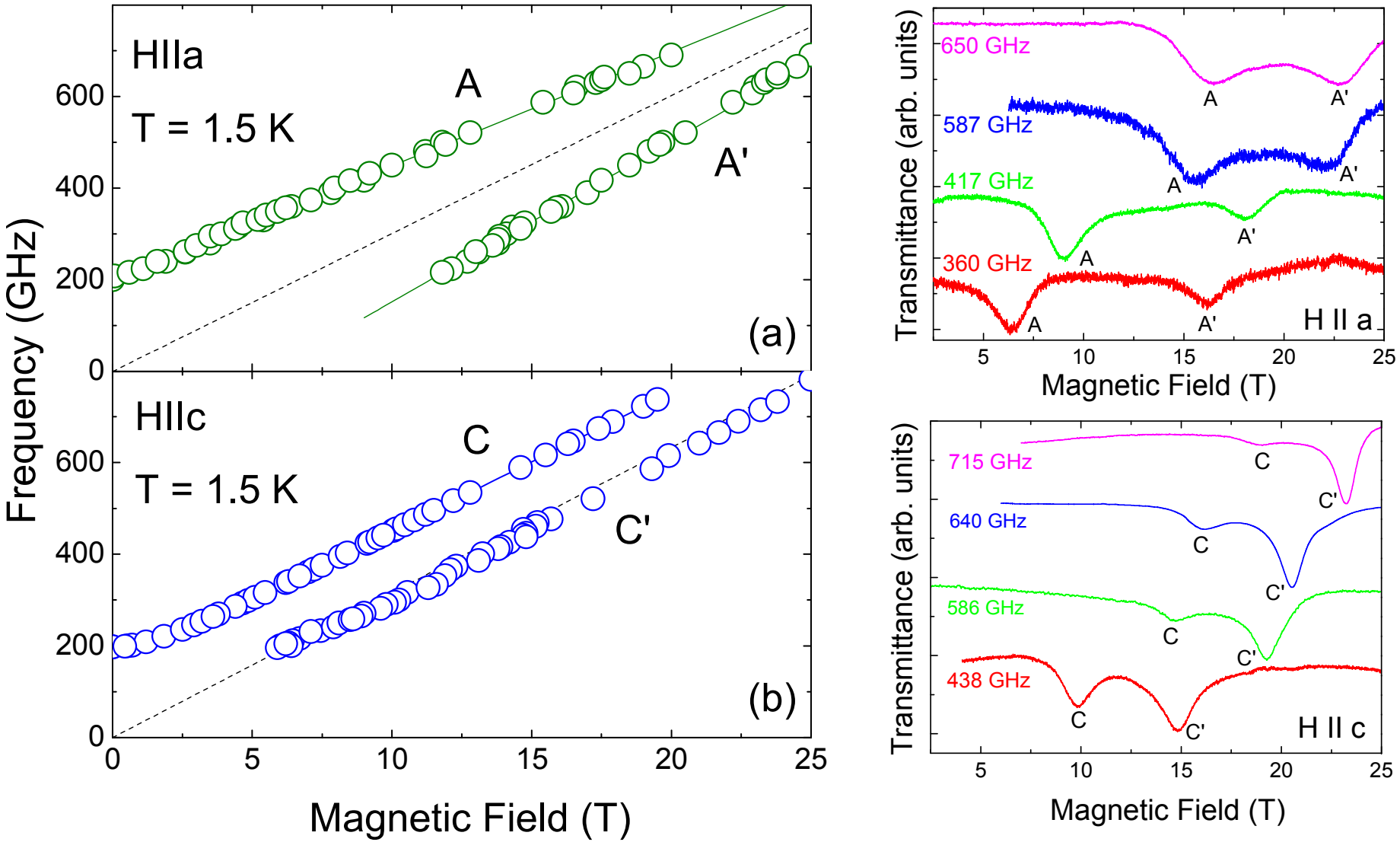
Calculations for triangular-lattice antifferomagnet (toy model)







Gap size: very good agreement between calculations and experimental results. Staggered DM interaction along the “chain” direction makes ESR mode observable.


HIIa
T = 1.5 K
A
A'
(a)
HIIc
T = 1.5 K
C
C'
(b)
Frequency (GHz)
Magnetic Field (T)
650 GHz
587 GHz
417 GHz
360 GHz
H II a
715 GHz
640 GHz
586 GHz
438 GHz
H II c
Transmittance (arb. units)
Magnetic Field (T)

Conclusions

- Based on the classical linear spin-wave theory approach applied above H_{sat}, exchange coupling parameters in Cs_2CuBr_4 and Cs_2CuCl_4 are accurately determined

- The approach has a broader impact and can be potentially used for any quantum magnet with reduced (e.g., by the staggered DM interaction) translational symmetry, resulting, as predicted, in emergence of a new exchange mode above H_{sat}.

- We show that the high-energy excitation spectrum is not significantly affected by the transition into the magnetically ordered state, indicating the co-existence of the magnetically-ordered ground state and excited states determined by short-range correlations.

- A pronounced zero-field gap is observed in the ESR excitation spectrum in Cs_2CuBr_4. The experimental data are compared with results of calculations for the spin-1/2 triangular lattice AFM toy model; indicating good agreement between the experiment and the calculations.

Open Problems

- Testing the proposed toy model for Cs_2CuBr_4 by inelastic neutron scattering experiments
- Looking for the modes of AFMR below T_N
- Including the DM interaction (staggered and uniform) in the calculations
- Calculating the excitation spectrum in magnetic field across remaining regions of the phase diagram

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