

Magnetic properties and spin dynamics in low-dimensional and frustrated magnets as probed by neutron scattering

New neutron-scattering results

$\alpha - \text{Cu}_2\text{V}_2\text{O}_7$ and $(\text{Rb}_{1-x}\text{Cs}_x)_2\text{Cu}_3\text{SnF}_{12}$

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MELODY 2026

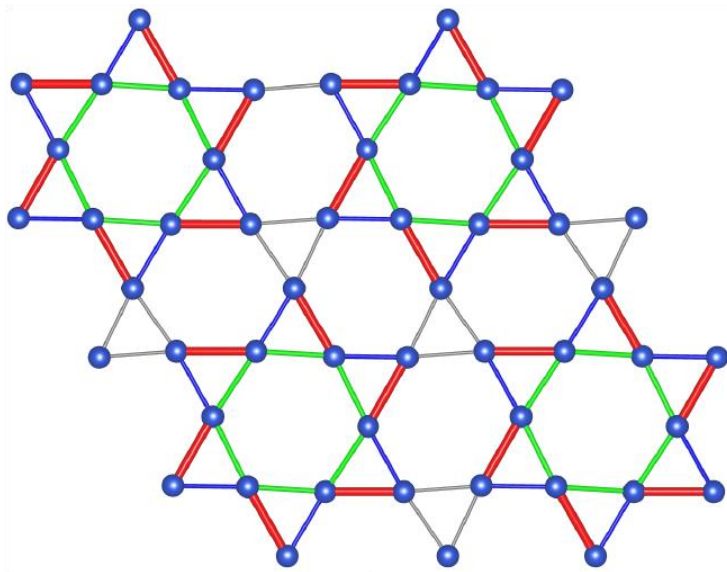
Workshop on Muon Science Technology and Industry

China Spallation Neutron Source (CSNS)

Rb/Cs kagome fluorides: singlets to magnetic order

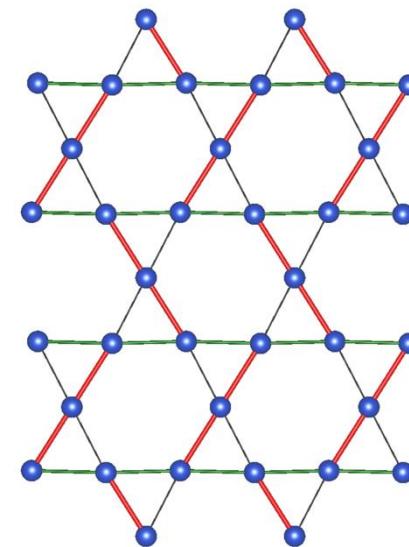
$(\text{Rb}_{1-x}\text{Cs}_x)_2\text{Cu}_3\text{SnF}_{12}$: $S = \frac{1}{2}$ kagome network

$\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$ ($x = 0$)



Pinwheel valence-bond solid
Spin-singlet ground state
Gap ≈ 2.35 meV

$\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ ($x = 1$)



Coplanar 120° order
 $T_n = 20.2$ K, $P2_1/n$
Negative vector chirality

Substitution changes exchange bonds without diluting the Cu network

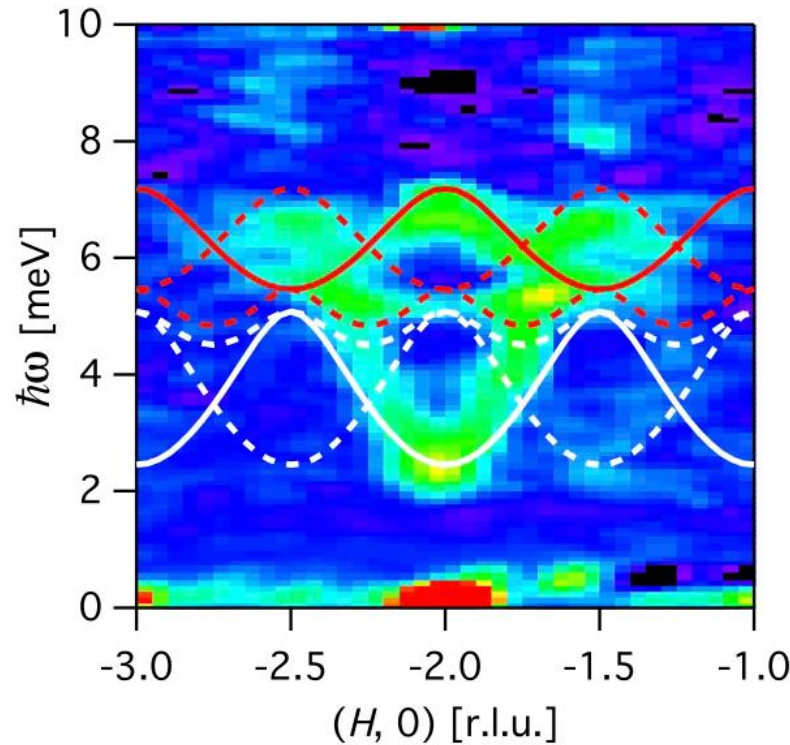
New results: $x = 0.2$ and $x = 0.5$ ($\text{RbCsCu}_3\text{SnF}_{12}$)

Rb: K. Matan et al., Nat. Phys. 6, 865 (2010);
Phys. Rev. B 89, 024414 (2014).

Cs: K. Matan et al., PRB 99, 224404 (2019);
Erratum: PRB 114, 139902(E) (2026).

Parent kagome spectra and spin Hamiltonians

$\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$ $x = 0$ · CNCS, SNS



Dispersive singlet–triplet excitations

Matan et al., Phys. Rev. B 89, 024414 (2014), Fig. 2(b)

$$\mathcal{H}_{\text{Rb}} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$

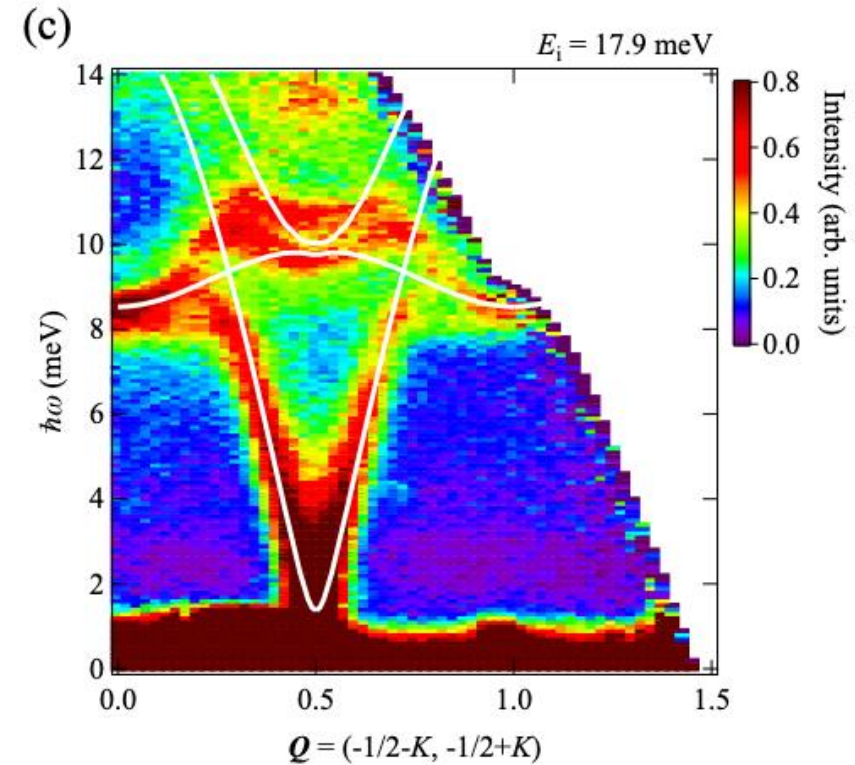
$$J_1 = 18.6 \text{ meV}, J_2/J_1 = 0.95$$

$$J_3/J_1 = 0.85, J_4/J_1 = 0.55$$

$$\text{Out-of-plane DM: } |D_{z_{ij}}| / J_{ij} = 0.18$$

Cs excitations: T. Ono et al., JPSJ 83, 043701 (2014); K. Matan et al., PRB 105, 134403 (2022).

$\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ $x = 1$ · 4SEASONS, J-PARC



Dispersive magnons in the ordered phase

Saito et al., Phys. Rev. B 105, 064424 (2022), Fig. 2(c)

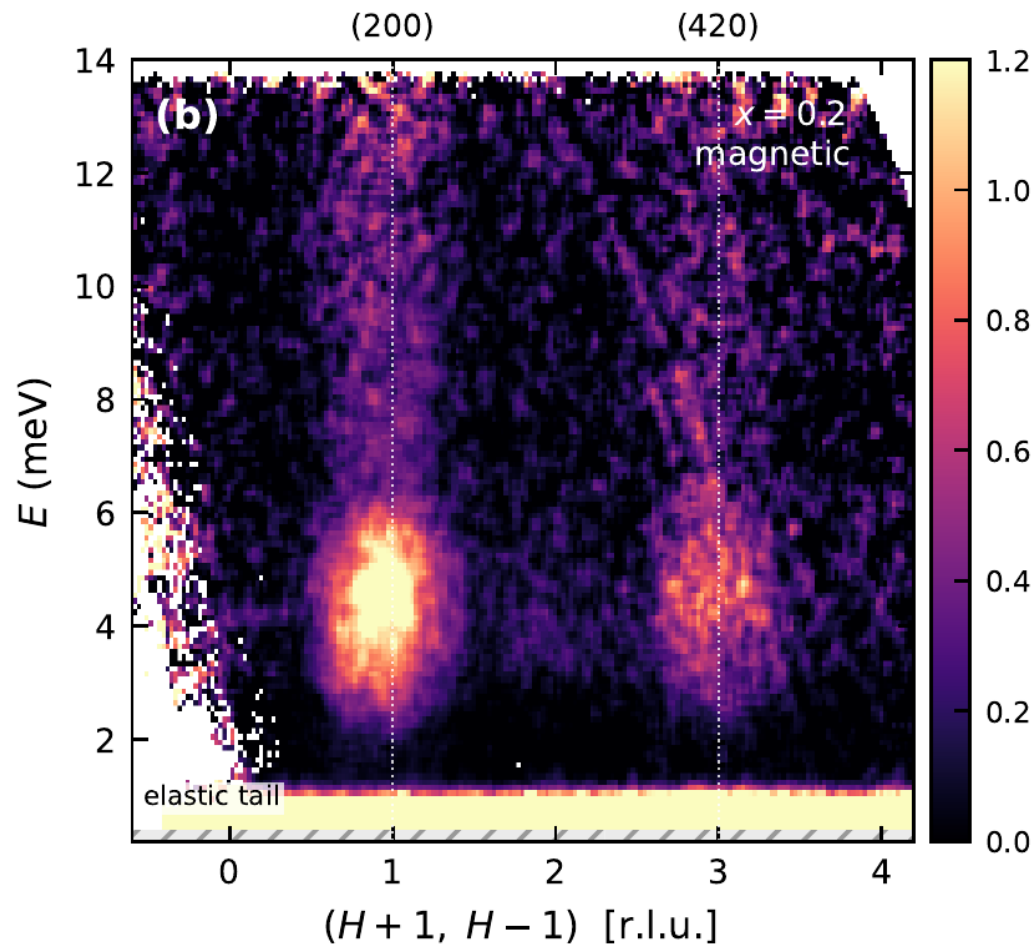
$$\mathcal{H}_{\text{Cs}} = J \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j + J' \sum_{\langle\langle ij \rangle\rangle} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij \rangle} \mathbf{D}_{ij} \cdot (\mathbf{S}_i \times \mathbf{S}_j)$$

$$J = 12.8 \text{ meV}, J'/J = -0.043$$

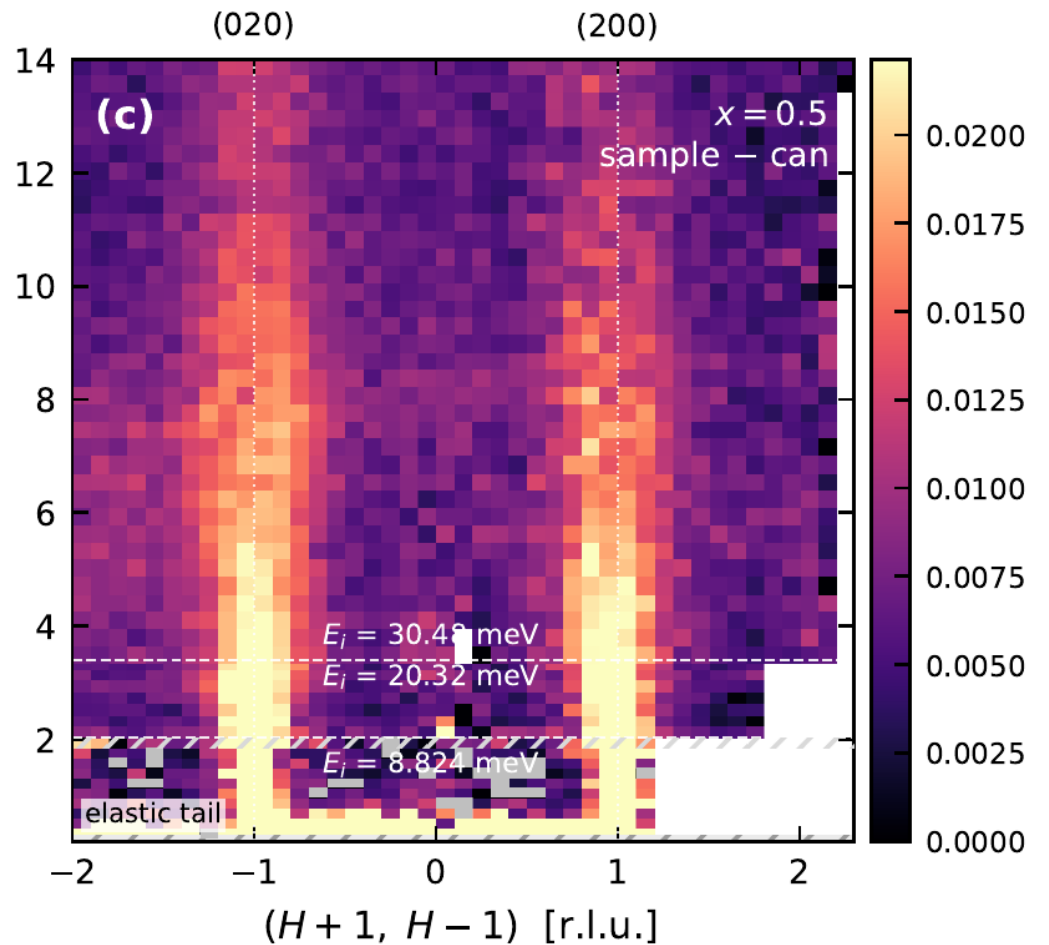
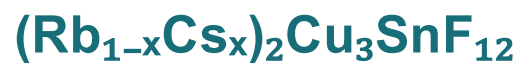
$$D_{\parallel}/J = 0.18, D_{\perp}/J = 0.062$$

Rb/Cs substitution destroys triplon coherence first

AMATERAS, J-PARC



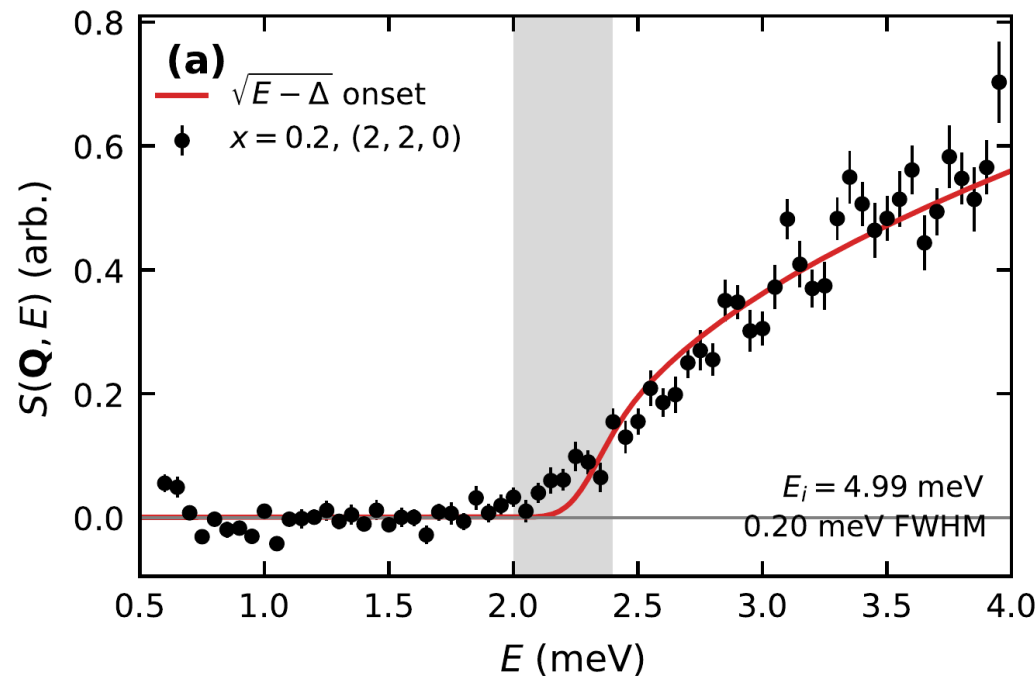
HRC, J-PARC



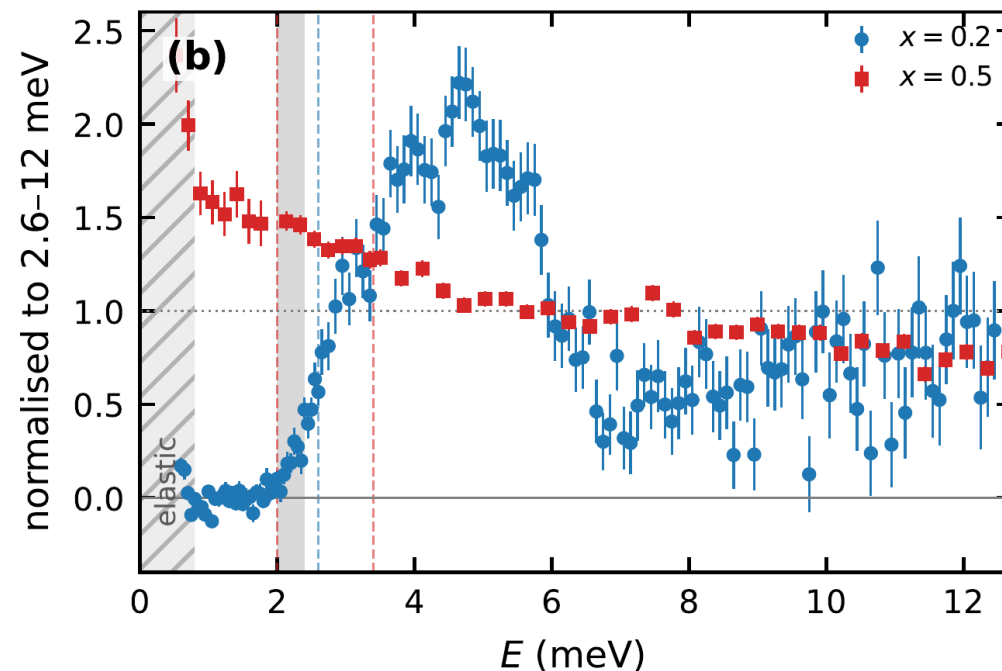
$x = 0.2$: non-dispersive rods with a surviving gap

$x = 0.5$: low-energy weight fills the gap region

The gap survives at $x = 0.2$ and fills near $x = 0.5$



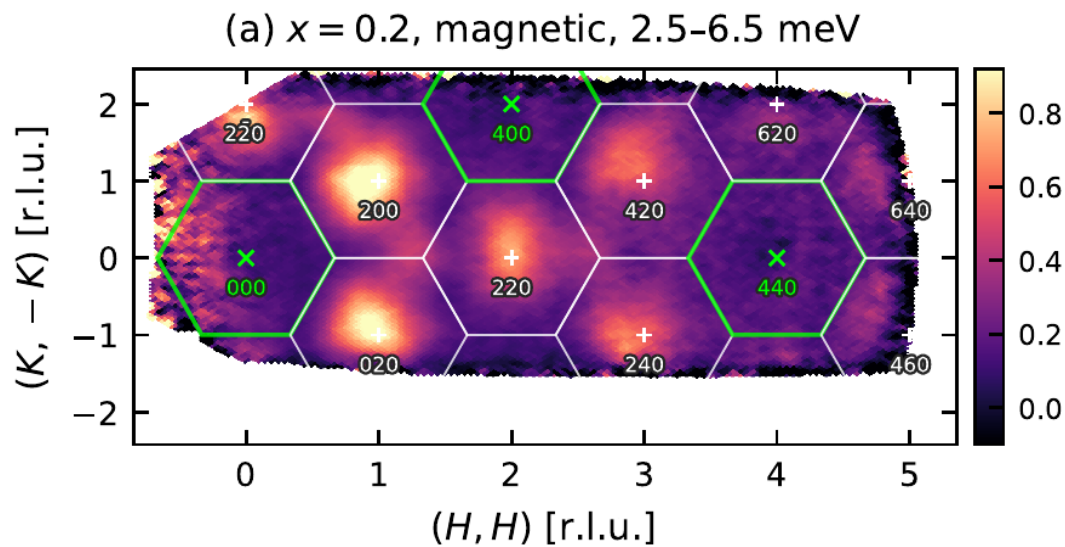
$x = 0.2$
Gap $\Delta = 2.0$ – 2.4 meV



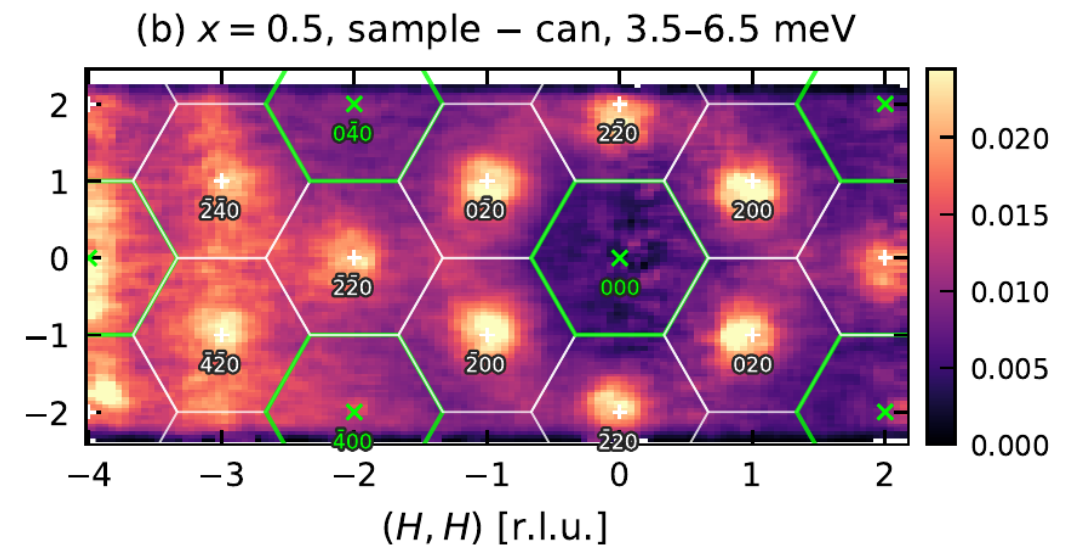
$x = 0.5$
No gap signature
Filled gap

Kagome selection-rule suppression survives

AMATERAS, J-PARC



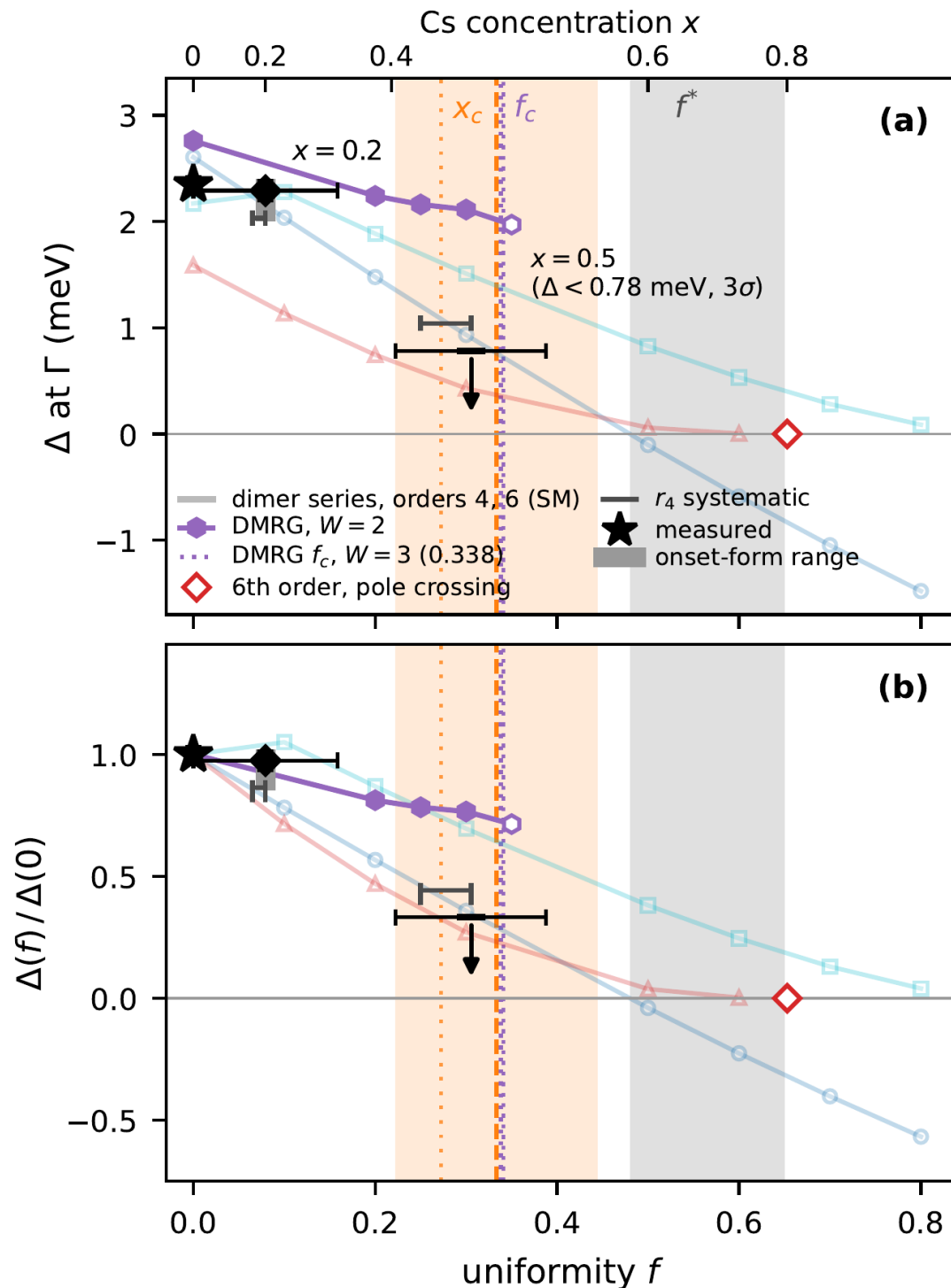
HRC, J-PARC



Strong suppression at $(4m, 4n)$ persists at both compositions
The gap-filling response retains the kagome momentum pattern

Parent-compound background: K. Matan et al., PRB 89, 024414 (2014); PRB 105, 134403 (2022).

Exchange uniformity and the loss of the singlet gap



Clean pinwheel model

DMRG: VBS branch ends near $f \approx 0.34$ on finite cylinders

x : Cs fraction

f : bond uniformity (0: parent, 1: uniform)

Susceptibility-based estimates:

$x = 0.2 \rightarrow f \approx 0.08$

$x = 0.5 \rightarrow f \approx 0.31$

Disorder-rounded collapse:

a working interpretation

Calculations remain unconverged

Susceptibility-derived x - f mapping:
 K. Katayama, N. Kurita and H. Tanaka,
 Phys. Rev. B 91, 214429 (2015), Fig. 3(b).

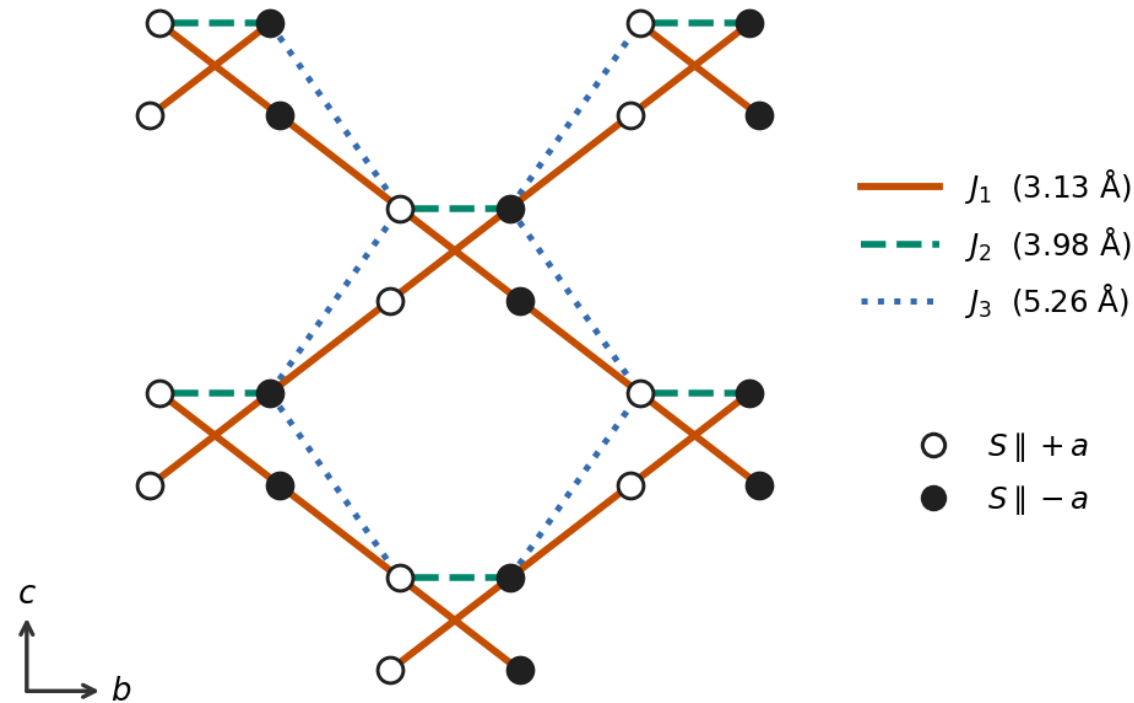
$\alpha\text{-Cu}_2\text{V}_2\text{O}_7$: polar antiferromagnet

Structure: G. Glathe et al., PRB 92, 024423 (2015); Erratum: PRB 95, 099902(E) (2017).

Polar domains: P. Piyawongwatthana et al., JPSJ 90, 025003 (2021); lattice: X. Hao et al., RPP 89, 018005 (2026).

(a) Exchange network: bc view

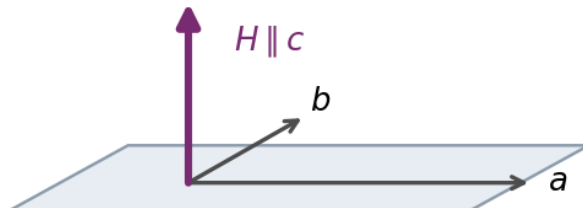
Slab: $0 \leq x \leq a/2$



(c) CNCS

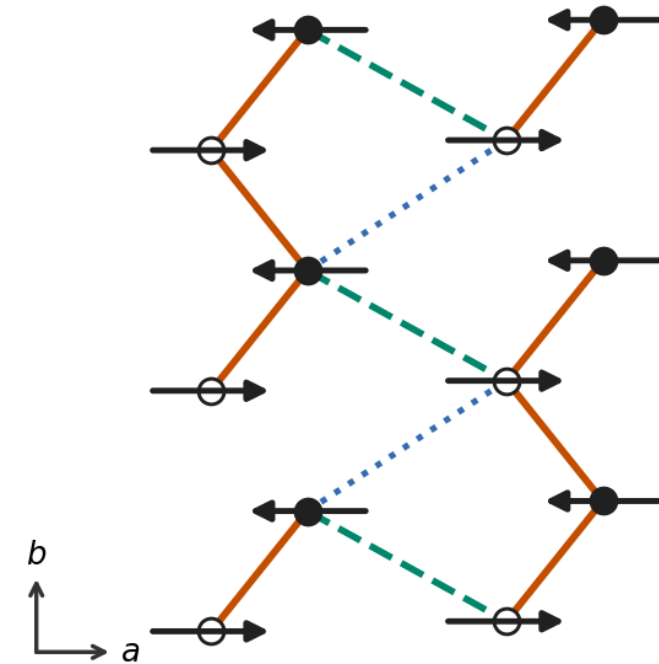
0–14 T

$H \perp S$



(b) Ordered moments: ab view

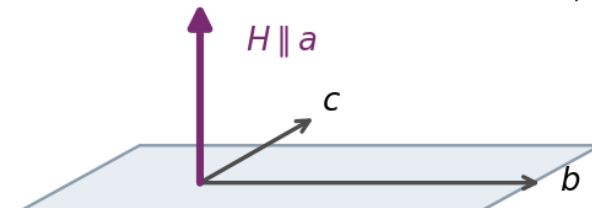
Slab: $0 \leq z \leq c$



(d) AMATERAS

0, ± 4 T

$H \parallel S$



$S = \frac{1}{2}$, polar $Fdd2$

$T_n = 33.4$ K

Dominant spins along a

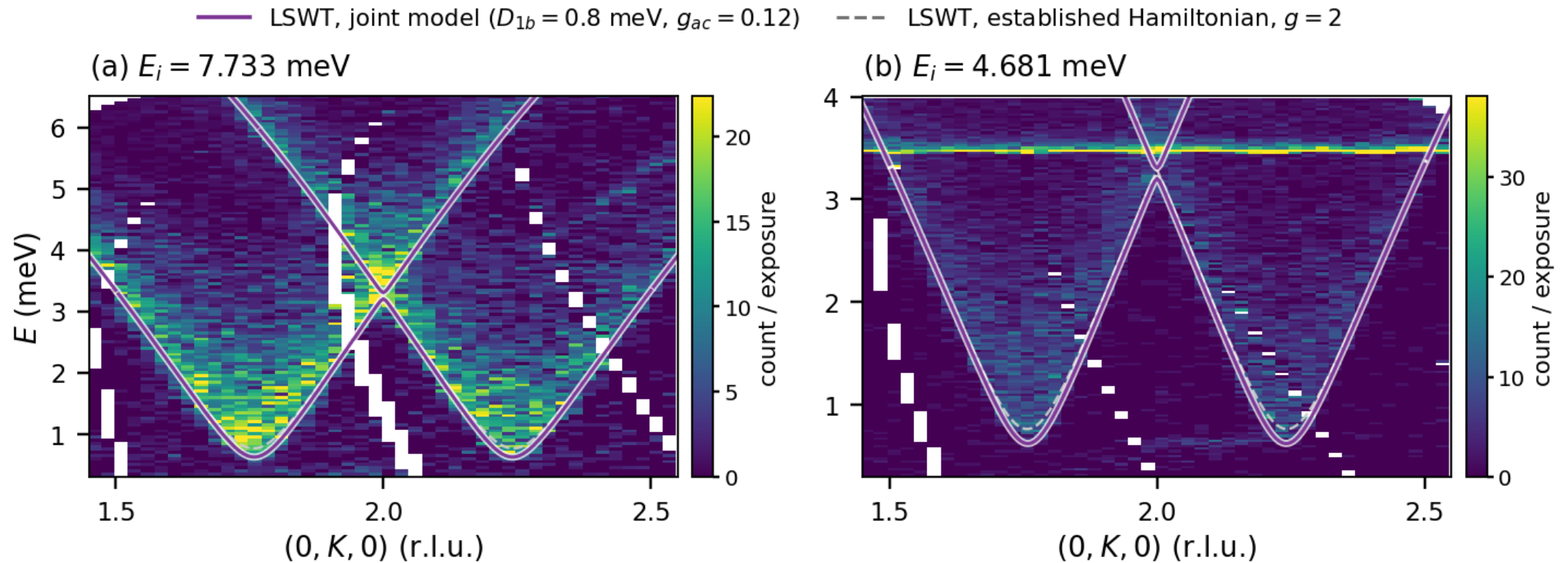
$H \parallel a$: branch reversal

$H \parallel c$: gap growth

Weak moment along c

$\approx 0.082 \mu_B$ per Cu

α -Cu₂V₂O₇: new AMATERAS zero-field spectra

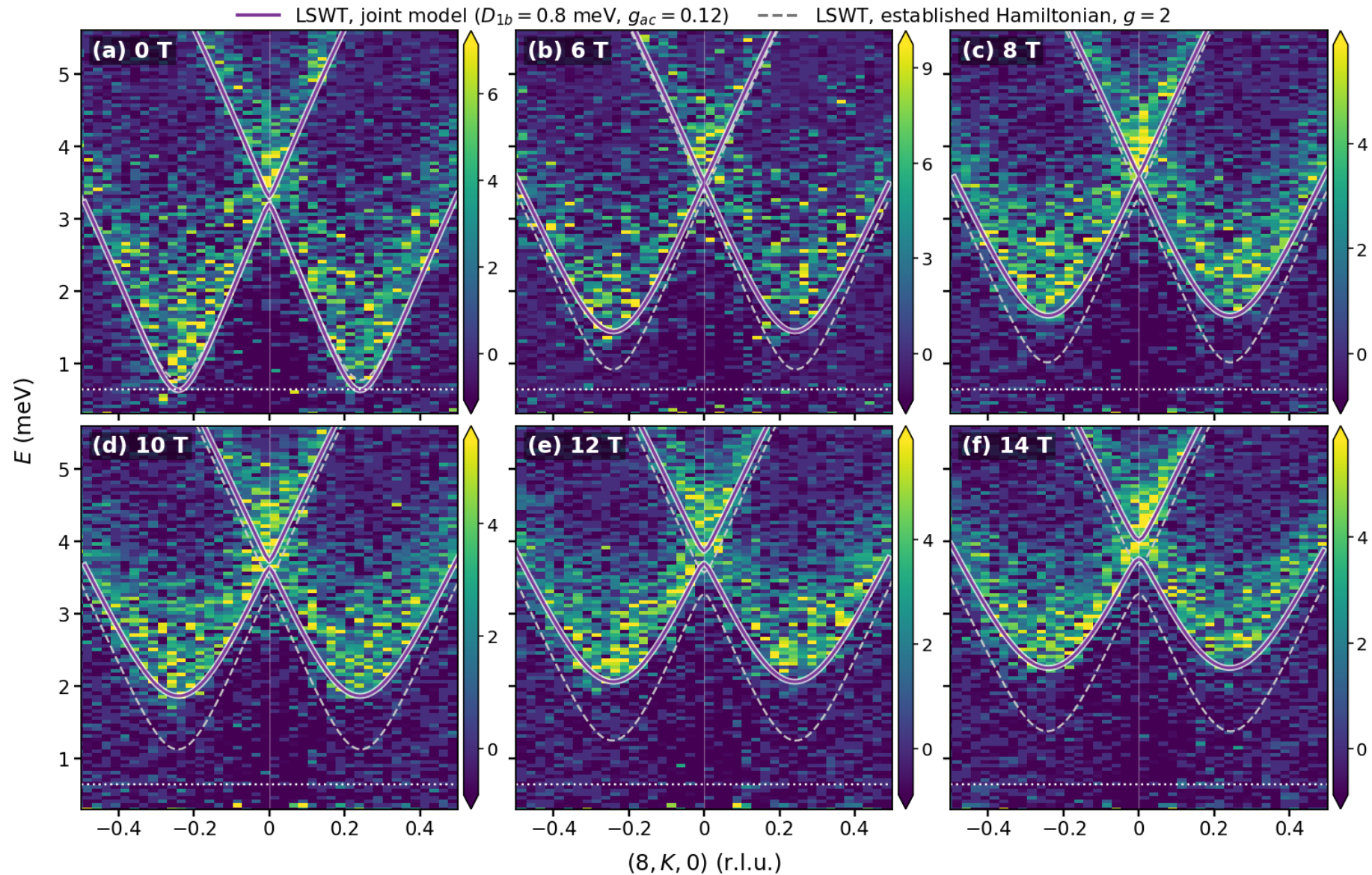


AMATERAS, 4 K, $H = 0$

Two displaced minima with a minimum-energy scale near 0.6 meV

H $\parallel$ c: contour maps show the gap growth to 14 T

CNCS, SNS



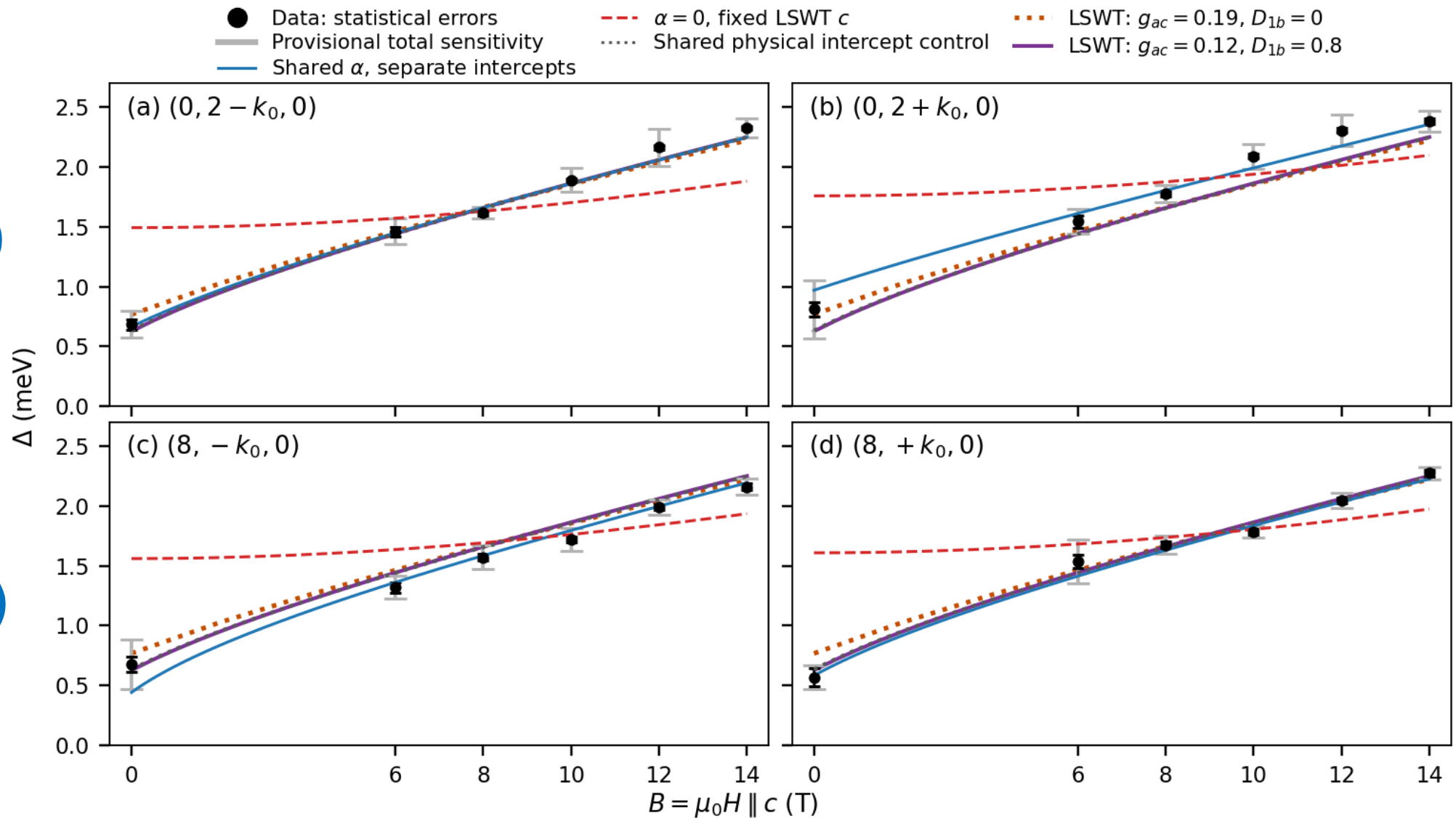
Colour bars: background-subtracted intensity (10^{-3} arb. units), separate scale for each panel

1.8 K: minimum gap around $(8,0,0)$ grows from ~ 0.6 to ~ 2.3 meV

Gap growth: quantitative comparison at four minima

(0,2,0)

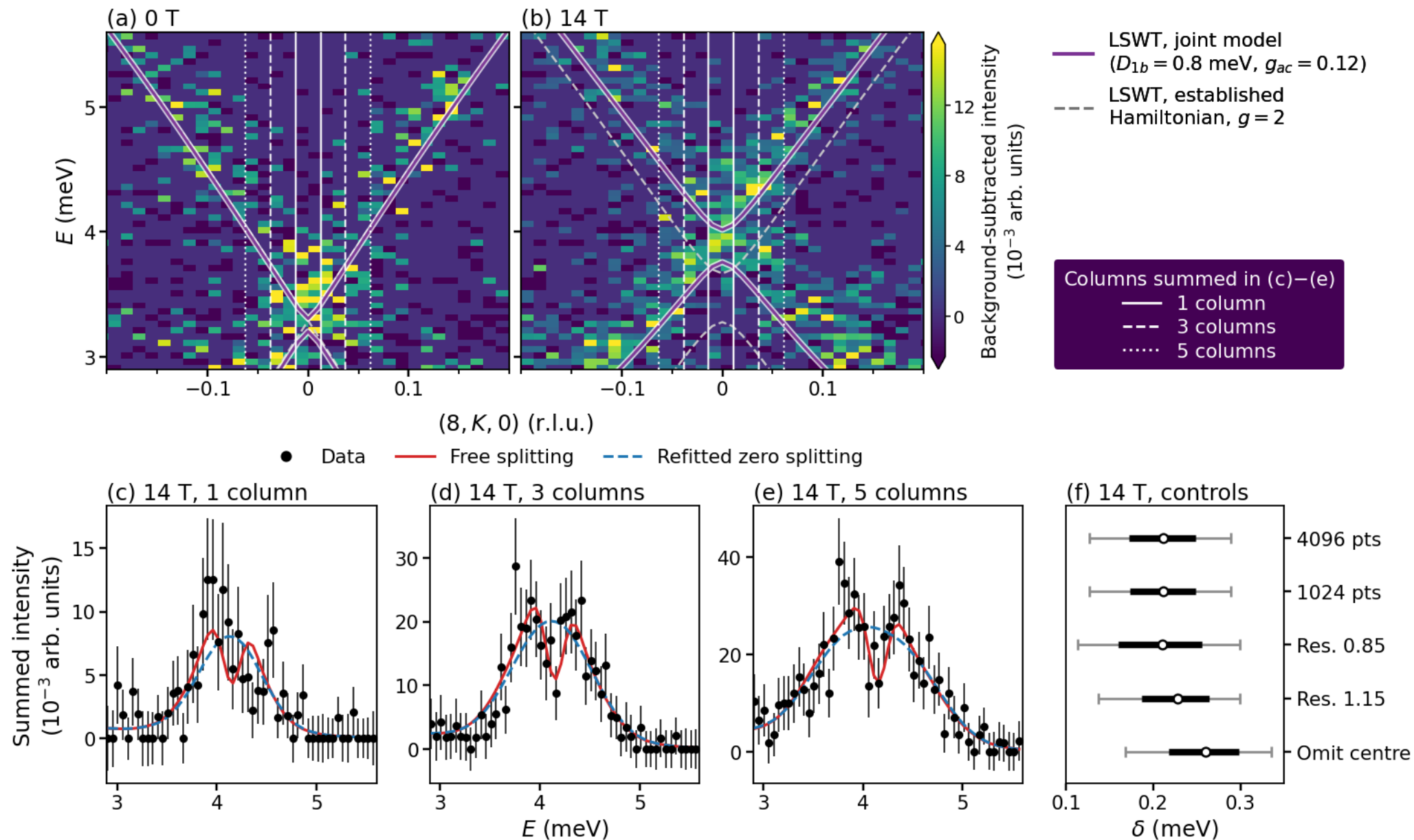
(8,0,0)



Four band minima show the same large increase

The established isotropic-Zeeman model underestimates the response

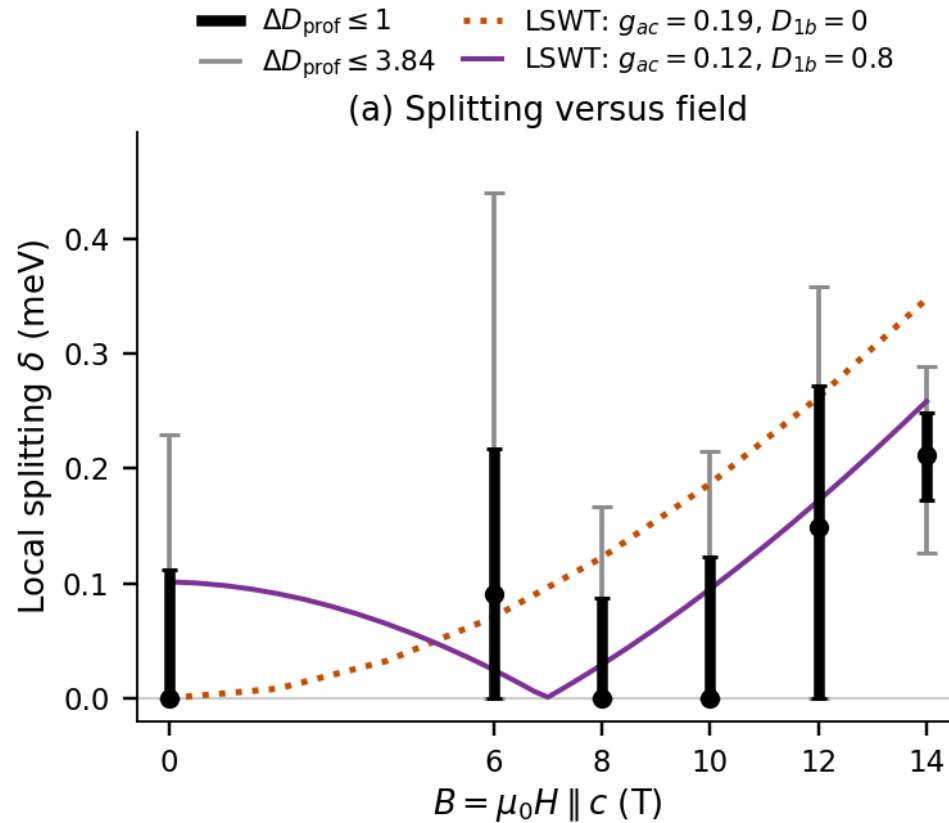
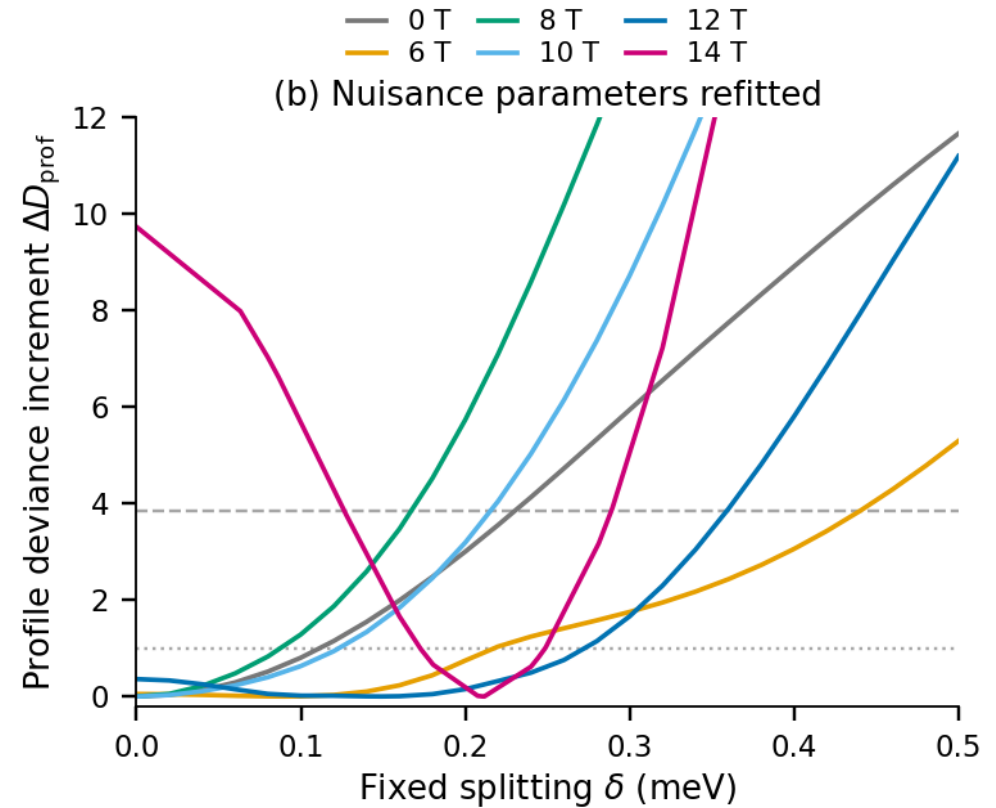
Zone-centre crossing: contour maps at 0 and 14 T



14 T: anti-crossing splitting ≈ 0.2 meV within the tested peak-profile model

Crossing splitting versus field

Nominal response: profile sets (uncalibrated); bootstrap-calibrated points at 0 and 14 T in the text



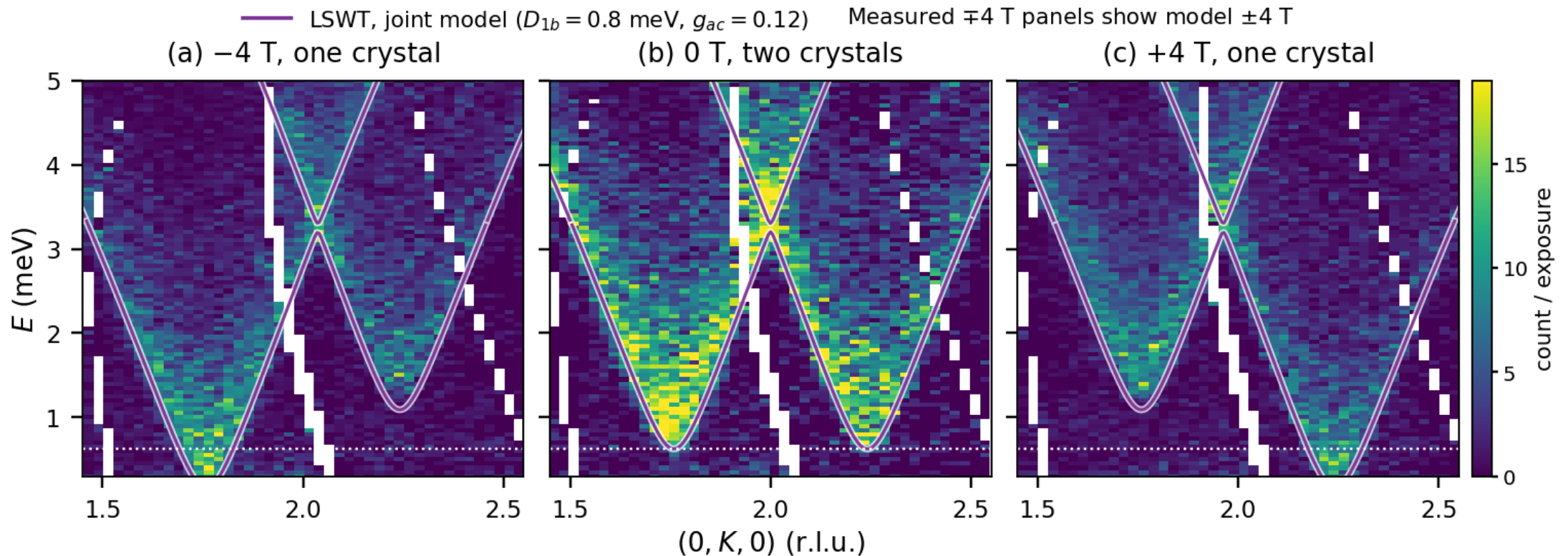
0 T: no detected splitting

14 T: splitting ≈ 0.2 meV within the tested peak-profile model

Minimum gap Δ and crossing splitting δ constrain different parts of the model

H || a: reversing the field interchanges the branches

AMATERAS, J-PARC



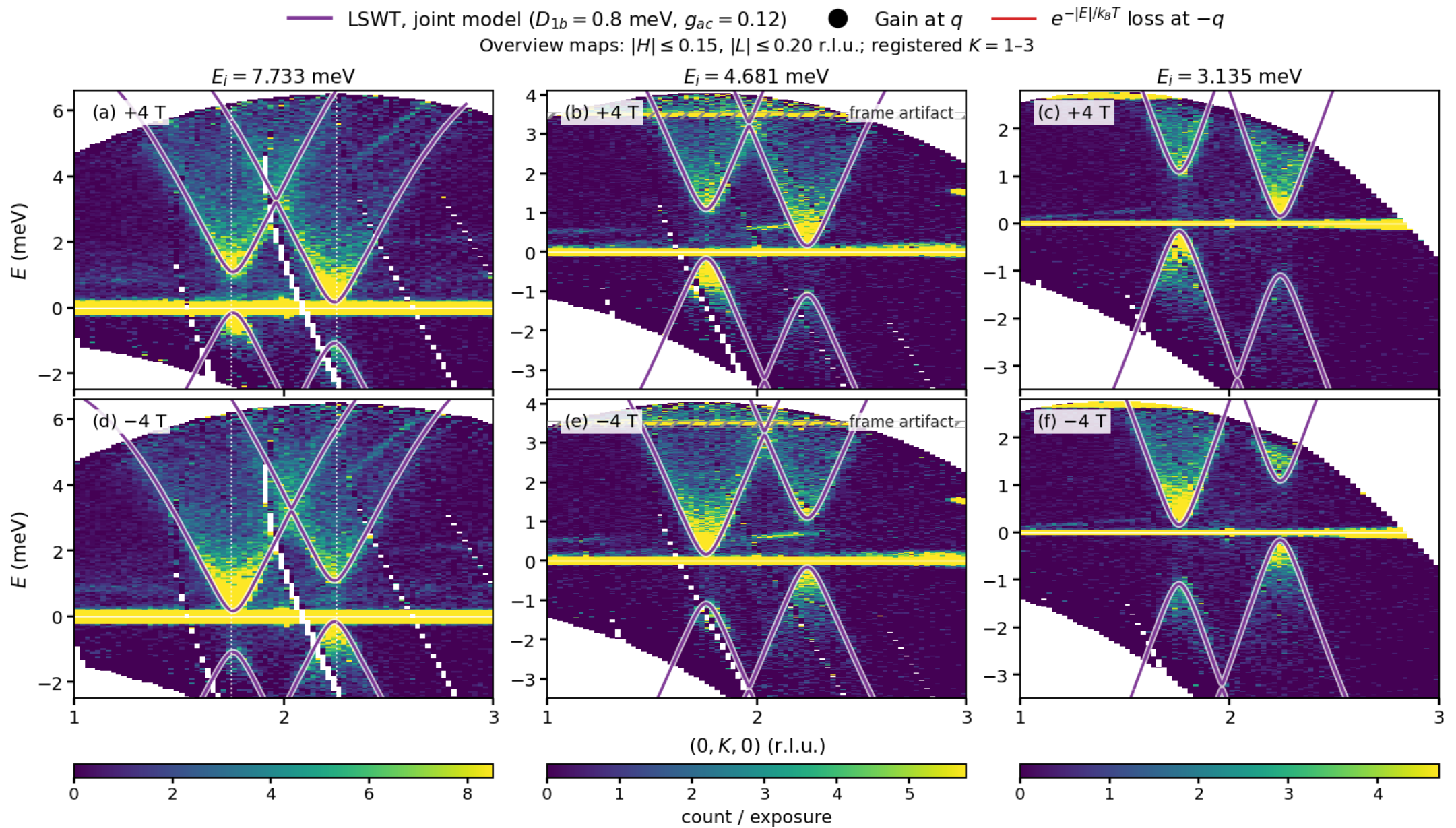
Opposite precession senses give opposite Zeeman shifts

The lower-branch gap remains unresolved

Nonreciprocal magnons: G. Ghatge et al., Phys. Rev. Lett. 119, 047201 (2017).

Related spin transport: Y. Shiomi et al., Phys. Rev. B 96, 180414(R) (2017).

Detailed balance: energy-gain and energy-loss maps



$$S_B(-\mathbf{Q}, -E) = e^{-\frac{E}{k_B T}} S_B(\mathbf{Q}, E)$$

Revised Hamiltonian: weak canting $\mathbf{D}_{1b}$ + staggered $\mathbf{g}_{ac}$

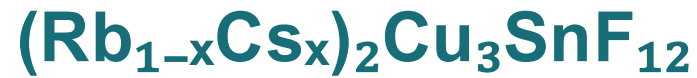
$$\mathcal{H} = \sum_{\langle ij \rangle} J_{ij} \mathbf{S}_i \cdot \mathbf{S}_j + \sum_{\langle ij \rangle_1} \mathbf{D}_1 \cdot (\mathbf{S}_i \times \mathbf{S}_j) \\ + G_1 \sum_{\langle ij \rangle_1} (S_i^a S_j^a - S_i^b S_j^b - S_i^c S_j^c) + \mu_B \sum_i \mathbf{H} \cdot \mathbf{g}_i \cdot \mathbf{S}_i$$

Original Model : $\mathbf{J}_1 = 2.67$, $\mathbf{J}_2 = 2.99$, $\mathbf{J}_3 = 5.42$, $\mathbf{D}_{1a} = 2.79$ meV; diagonal $\mathbf{g} = 2$

Added and refined parameters : $\mathbf{D}_{1b} = 0.80$ meV $\mathbf{g}_{ac} = \pm 0.120$ $\mathbf{G}_1 = 0.267$ meV

Observable	Joint model	Experiment	Comparison
Zero-field gap Δ_0 (meV)	0.625	0.64 ± 0.07	Consistent
Gap-growth α (meV ² /T)	0.241	0.236 ± 0.010	Consistent
0 T crossing δ (meV)	0.101	No detected splitting	Allowed by 0 T test
14 T crossing δ (meV)	0.259	$0.212 (+0.036/-0.039)$	Broader profile range
Moment (μ_B/Cu)	0.079 (harmonic)	0.082 ± 0.001	Close to measured value

Conclusions



Loss of coherent triplons precedes gap filling
The kagome momentum pattern survives



Large transverse-field ($H \parallel c$) gap growth
Crossing spectra constrain canting and Zeeman coupling jointly

The microscopic origin of the effective coupling remains open

<p>Sample synthesis</p> <p>Thermodynamics property and neutron scattering measurements</p>	Mahidol University	Pakornsak Saesun, Chairote Piyakulworawat
	Phranakhon Rajabhat University	Ganatee Gitgeatpong
	Tokyo Institute of Technology, Japan	H. Tanaka , T. Ono , N. Kurita, P. Chanlert
	ISSP, University of Tokyo, Japan	P. Piyawongwatthana T. J. Sato , M. Tokunaga, A. Miyake
	National Taiwan University, Taiwan	W.-T Chen, F.C. Chou
	NIST Center for Neutron Research	Y. Zhao, N. Butch, Y. Qiu, L.W. Harriger
	Spallation Neutron Source, ORNL	A. Podlesnyak, A. T. Savici, C. Schmitt
	J-PARC	M. Kofu, N. Murai, S. Ohira- Kawamura, Kenji Nakajima
	Bragg Institute, ANSTO, Australia	M. Avdeev, R. Splitz
DFT	Mahidol University	M. Suewattana
	College of William and Mary, USA	Shiwei Zhang

Related papers: kagome fluorides

Pinwheel valence-bond solid and triplet excitations

K. Matan et al.
Nat. Phys. 6, 865 (2010)
[doi: 10.1038/nphys1761](https://doi.org/10.1038/nphys1761)

Ghost modes and continuum scattering in $\text{Rb}_2\text{Cu}_3\text{SnF}_{12}$

K. Matan et al.
Phys. Rev. B 89, 024414 (2014)
[doi: 10.1103/PhysRevB.89.024414](https://doi.org/10.1103/PhysRevB.89.024414)

Quantum renormalization of $\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ excitations

T. Ono et al.
J. Phys. Soc. Jpn. 83, 043701 (2014)
[doi: 10.7566/JPSJ.83.043701](https://doi.org/10.7566/JPSJ.83.043701)

$\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$: structure and high-field magnetization

K. Matan et al.
Phys. Rev. B 99, 224404 (2019)
[doi: 10.1103/PhysRevB.99.224404](https://doi.org/10.1103/PhysRevB.99.224404)

Breakdown of LSWT and spinon bound states

K. Matan et al.
Phys. Rev. B 105, 134403 (2022)
[doi: 10.1103/PhysRevB.105.134403](https://doi.org/10.1103/PhysRevB.105.134403)

$\text{Cs}_2\text{Cu}_3\text{SnF}_{12}$ magnetic-structure correction

K. Matan et al.
Phys. Rev. B 114, 139902(E) (2026)
[doi: 10.1103/35kb-w65h](https://doi.org/10.1103/35kb-w65h)

Susceptibility and the substitution-driven transition

K. Katayama, N. Kurita and H. Tanaka
Phys. Rev. B 91, 214429 (2015)
[doi: 10.1103/PhysRevB.91.214429](https://doi.org/10.1103/PhysRevB.91.214429)

Related papers: copper pyrovanadate

α -Cu₂V₂O₇: magnetic structure and DM interaction

G. Gitgeatpong et al.
Phys. Rev. B 92, 024423 (2015)
[doi: 10.1103/PhysRevB.92.024423](https://doi.org/10.1103/PhysRevB.92.024423)

Erratum: symmetry allowance for next-neighbour DM

G. Gitgeatpong et al.
Phys. Rev. B 95, 099902(E) (2017)
[doi: 10.1103/PhysRevB.95.099902](https://doi.org/10.1103/PhysRevB.95.099902)

Nonreciprocal magnons and symmetry breaking

G. Gitgeatpong et al.
Phys. Rev. Lett. 119, 047201 (2017)
[doi: 10.1103/PhysRevLett.119.047201](https://doi.org/10.1103/PhysRevLett.119.047201)

High-field magnetization and magnetic phase diagram

G. Gitgeatpong et al.
Phys. Rev. B 95, 245119 (2017)
[doi: 10.1103/PhysRevB.95.245119](https://doi.org/10.1103/PhysRevB.95.245119)

Formation of a single polar domain in α -Cu₂V₂O₇

P. Piyawongwatthana et al.
J. Phys. Soc. Jpn. 90, 025003 (2021)
[doi: 10.7566/JPSJ.90.025003](https://doi.org/10.7566/JPSJ.90.025003)

Spin Seebeck effect in polar α -Cu₂V₂O₇

Y. Shiomi et al.
Phys. Rev. B 96, 180414(R) (2017)
[doi: 10.1103/PhysRevB.96.180414](https://doi.org/10.1103/PhysRevB.96.180414)

Review: nonreciprocal magnons

T. J. Sato and K. Matan
J. Phys. Soc. Jpn. 88, 081007 (2019)
[doi: 10.7566/JPSJ.88.081007](https://doi.org/10.7566/JPSJ.88.081007)

Jahn–Teller distortion and negative thermal expansion

X. Hao et al.
Rep. Prog. Phys. 89, 018005 (2026)
[doi: 10.1088/1361-6633/ae3853](https://doi.org/10.1088/1361-6633/ae3853)

Related β phase: Zn-doped copper pyrovanadate

G. Gitgeatpong et al.
Phys. Rev. B 106, 214438 (2022)
[doi: 10.1103/PhysRevB.106.214438](https://doi.org/10.1103/PhysRevB.106.214438)