# Quantum interaction effects in a triangular spin-1/2 antiferromagnet

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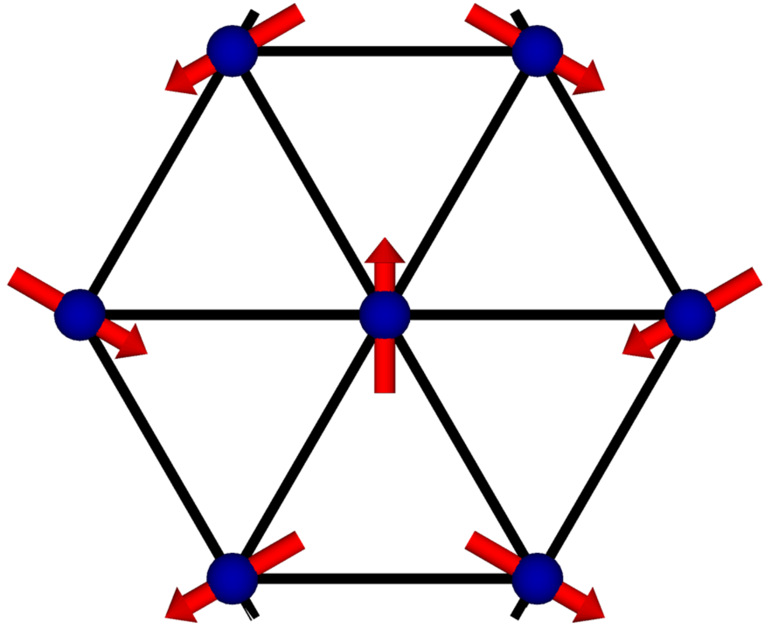
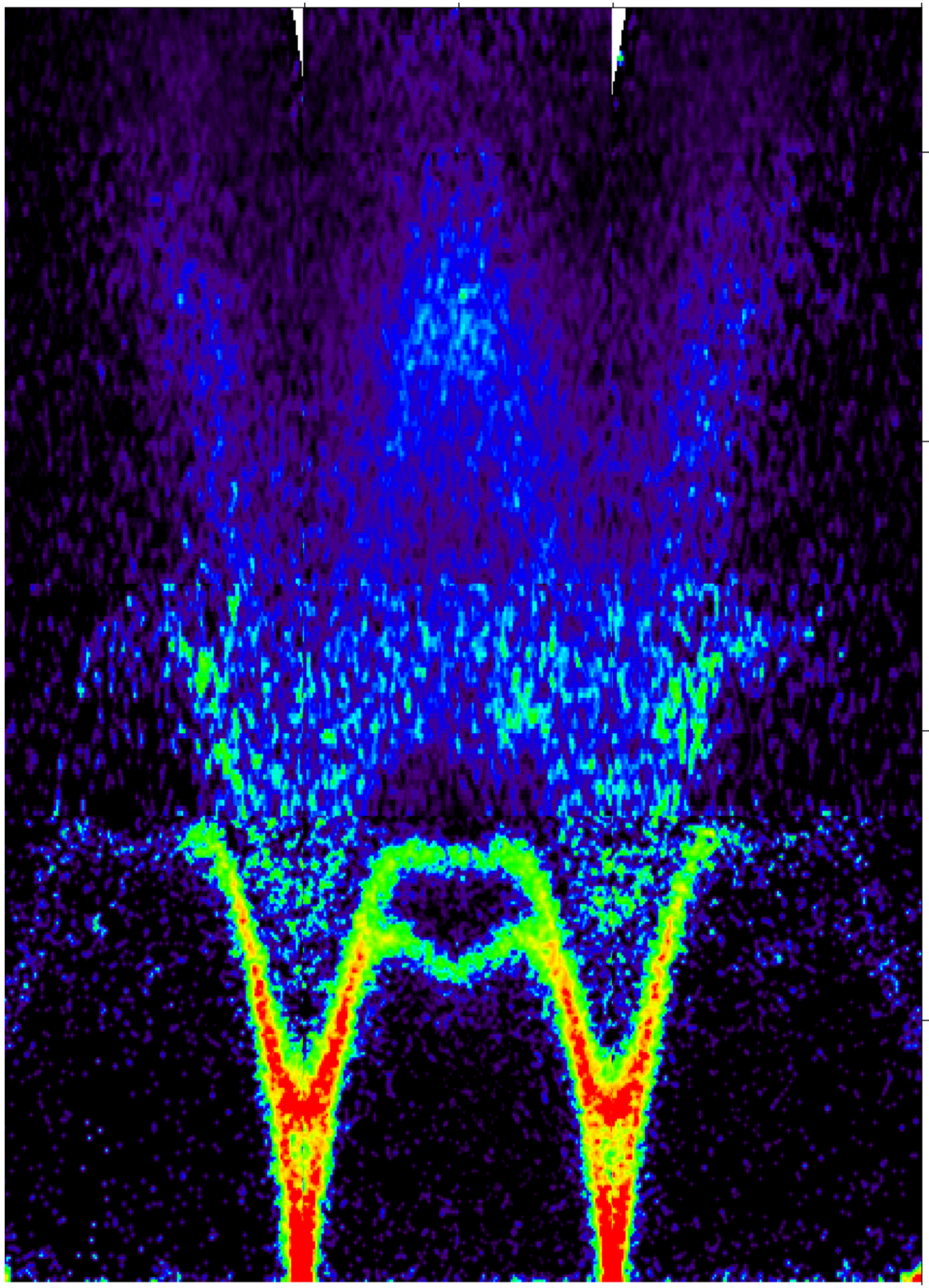
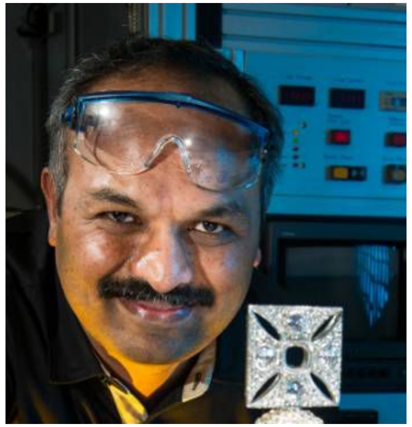
Oxford





*KITP, Correlated20*

# Collaborators



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Neutron Scattering on LET @ ISIS

European Research Council
Established by the European Commission

EPSRC
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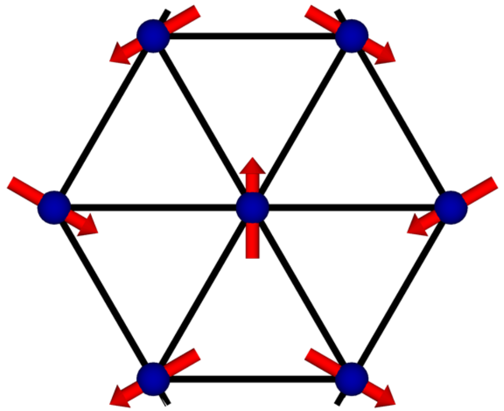
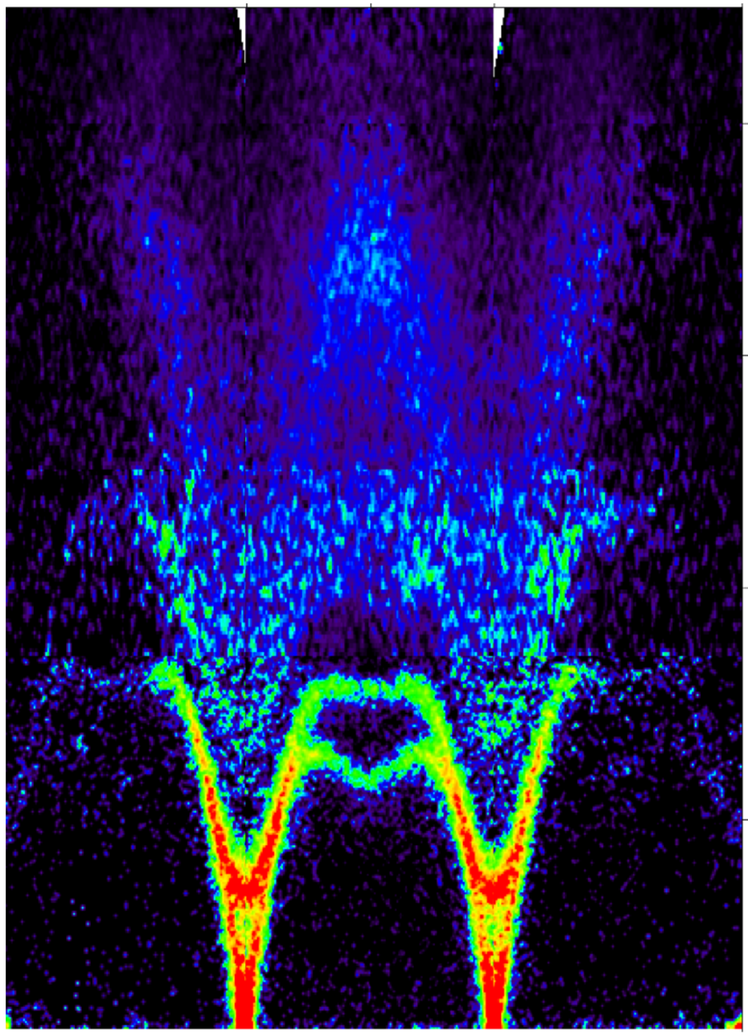
# Outline

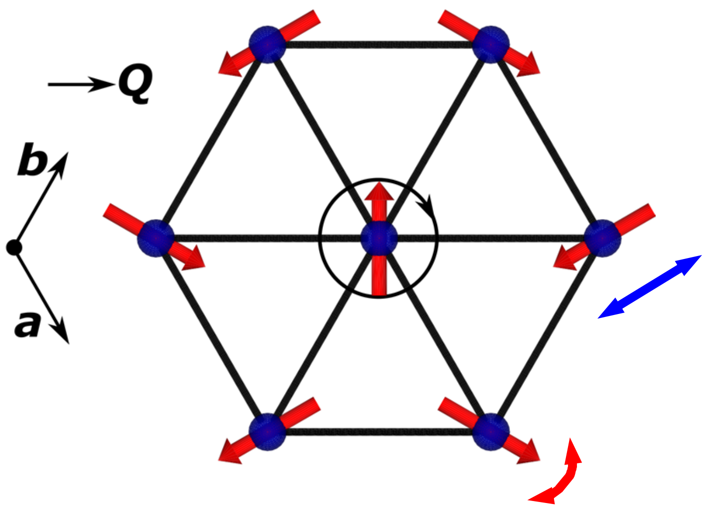


1) Triangular Heisenberg AFM – open questions on spin dynamics
2) $Ba_3CoSb_2O_9$:  $Co^{2+}$  effective $S = 1/2$ and 120° Nèel order
3) Overview of excitation spectrum
4) Magnon dispersions: renormalizations and soft modes
5) Why magnons do not decay?
6) High-energy continuum vs two-magnon excitations



*D. Macdougal,.. R.C, Phys. Rev. B 102, 064421 (2020)*

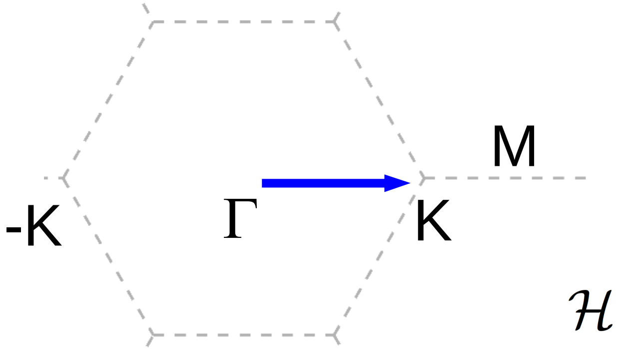
# Triangular Heisenberg antiferromagnet: spin-wave modes

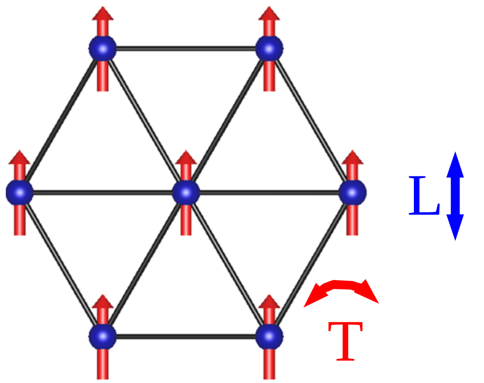



- spontaneous non-collinear 120° Nèel order with spin reduction $\Delta S$~0.3

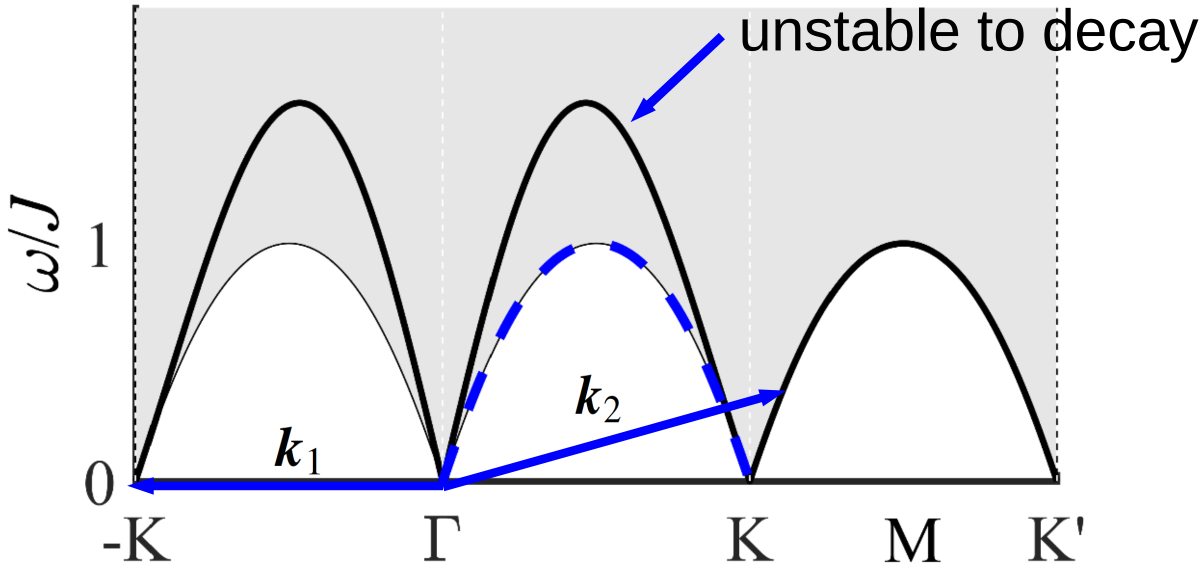
- single propagation vector $\boldsymbol{Q}$ = (1/3,1/3)

**local (rotating) frame**




$\mathcal{H}$ couples L & T components



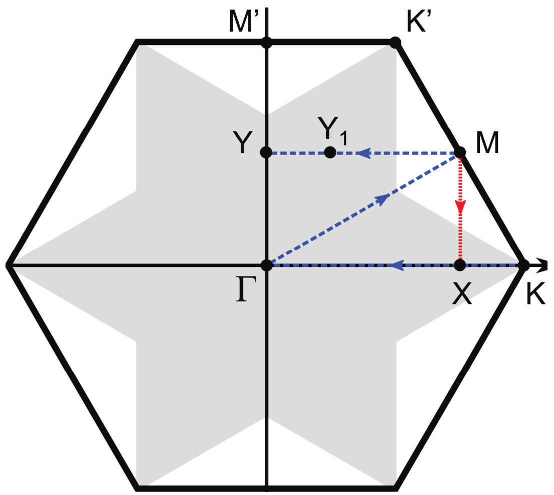

- two-magnon phase space

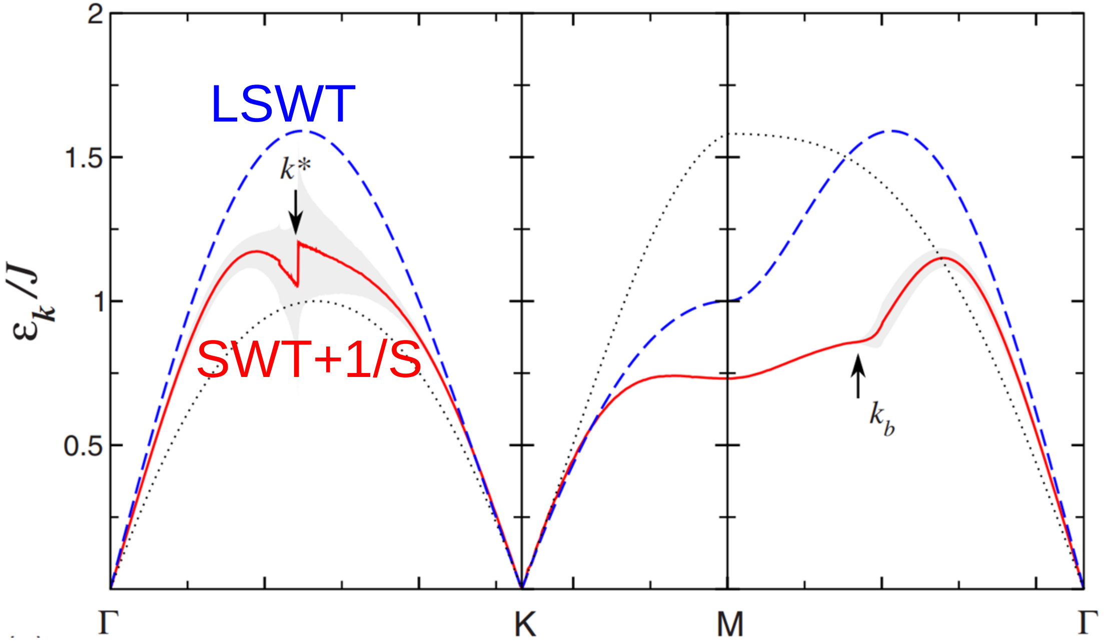
$\boldsymbol{k} = \boldsymbol{k}_1 + \boldsymbol{k}_2$

$\omega(\boldsymbol{k}) = \omega(\boldsymbol{k}_1) + \omega(\boldsymbol{k}_2)$

- in lab frame see ω($\boldsymbol{k}$), ω($\boldsymbol{k}\pm\boldsymbol{Q}$)

# Triangular Heisenberg antiferromagnet: spin-wave approaches



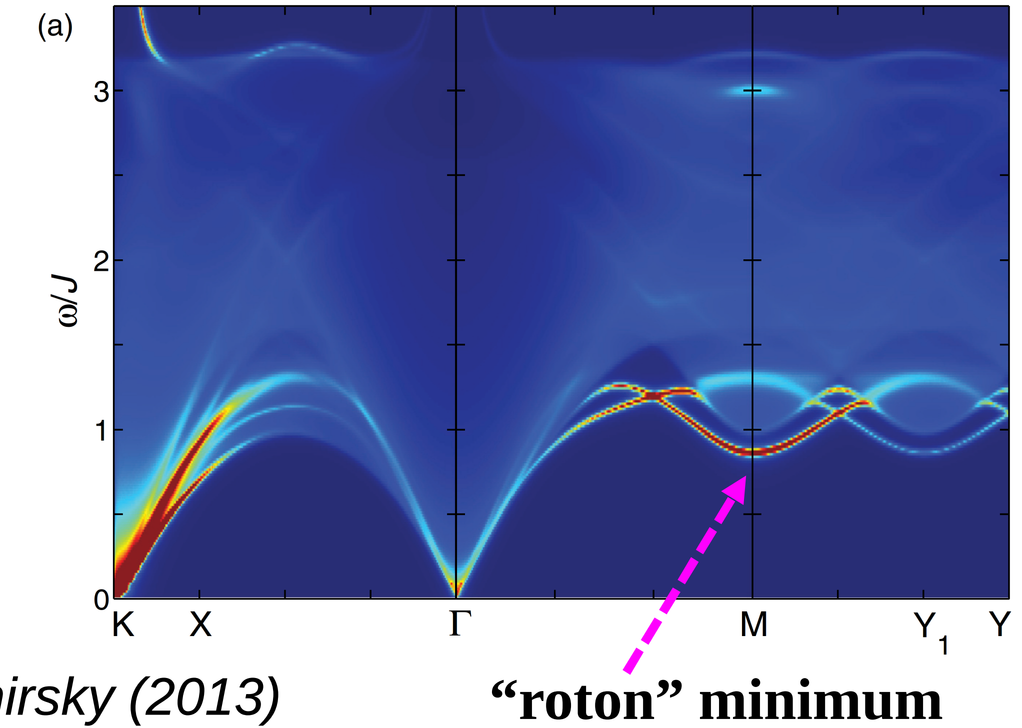

*Chernyshev, Zhitomirsky (2009)*
*Starykh,Chubukov… (2006)*

spin-wave approaches predict

- extended regions of one -> two magnon decays

- **downward dispersion renormalization**
(also found by series expansions)

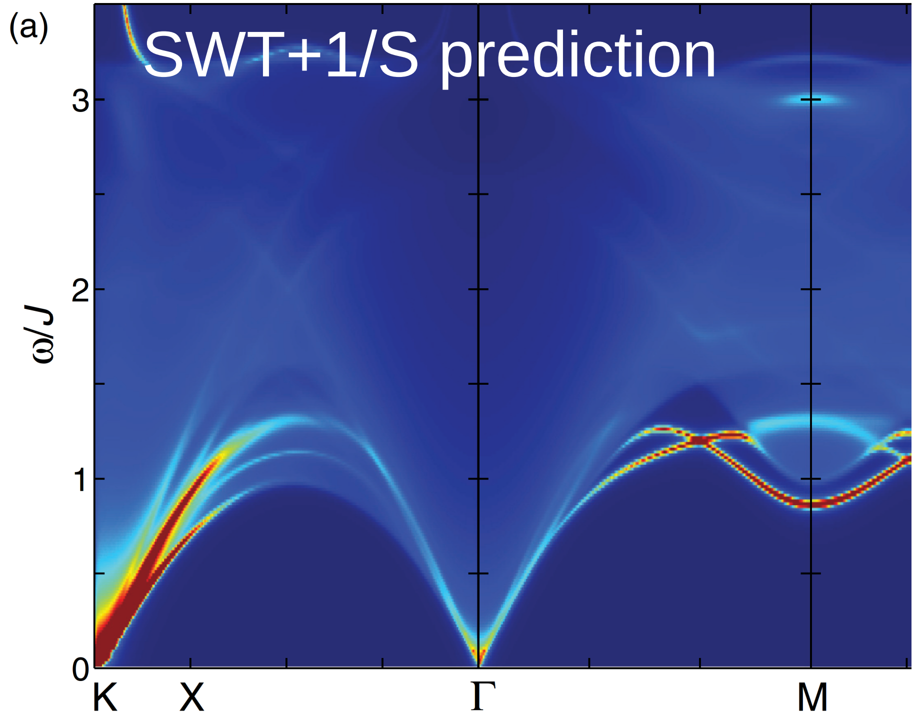
- roton **soft mode** at M




*Mourigal, Fuhrman, Chernyshev, Zhitomirsky (2013)*

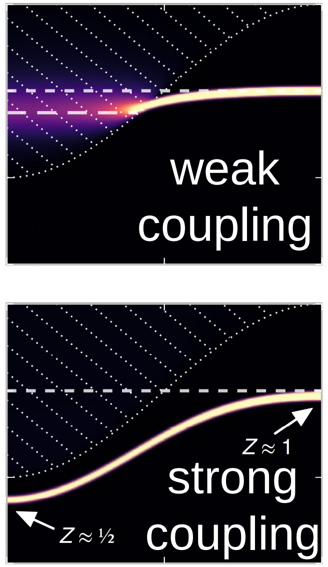
# Triangular Heisenberg antiferromagnet  - open questions

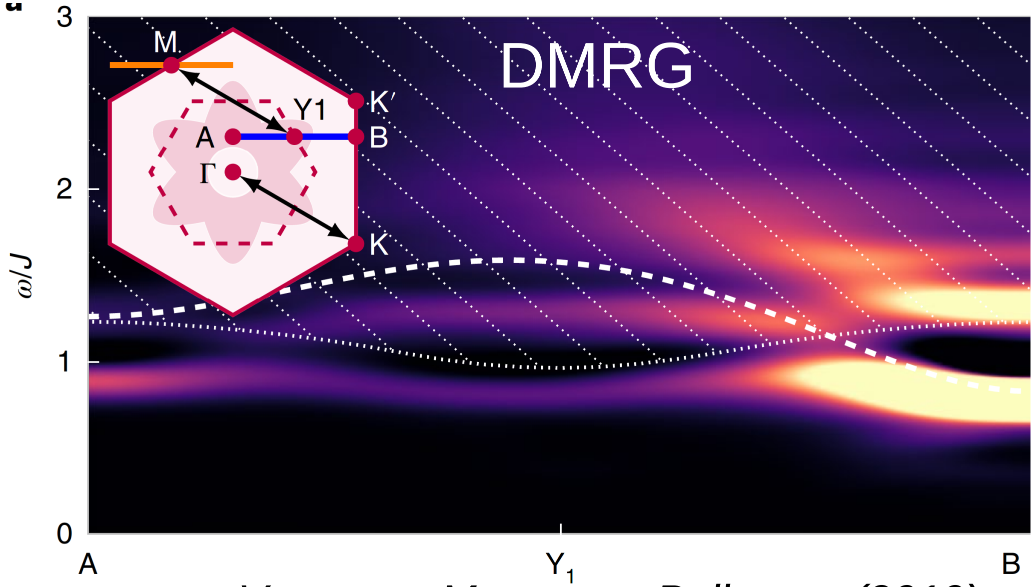
- do magnons decay ?
- can dispersion be described by SWT?
- nature of high-energy excitations, multi-spinwaves?



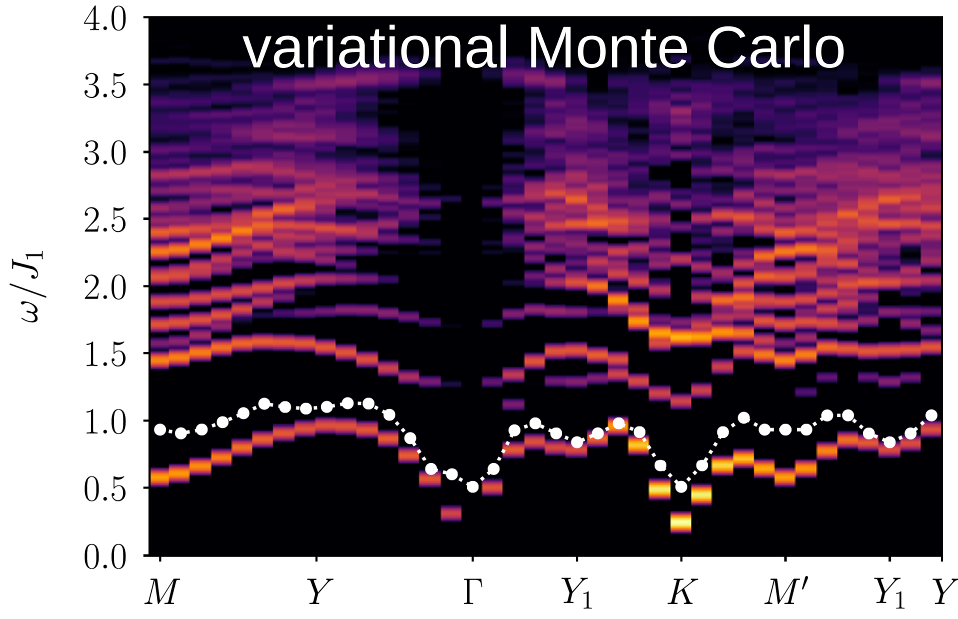

Mourigal, Fuhrman, Chernyshev, Zhitomirsky (2013)

- DMRG near HB limit find no decay, variational Monte Carlo also



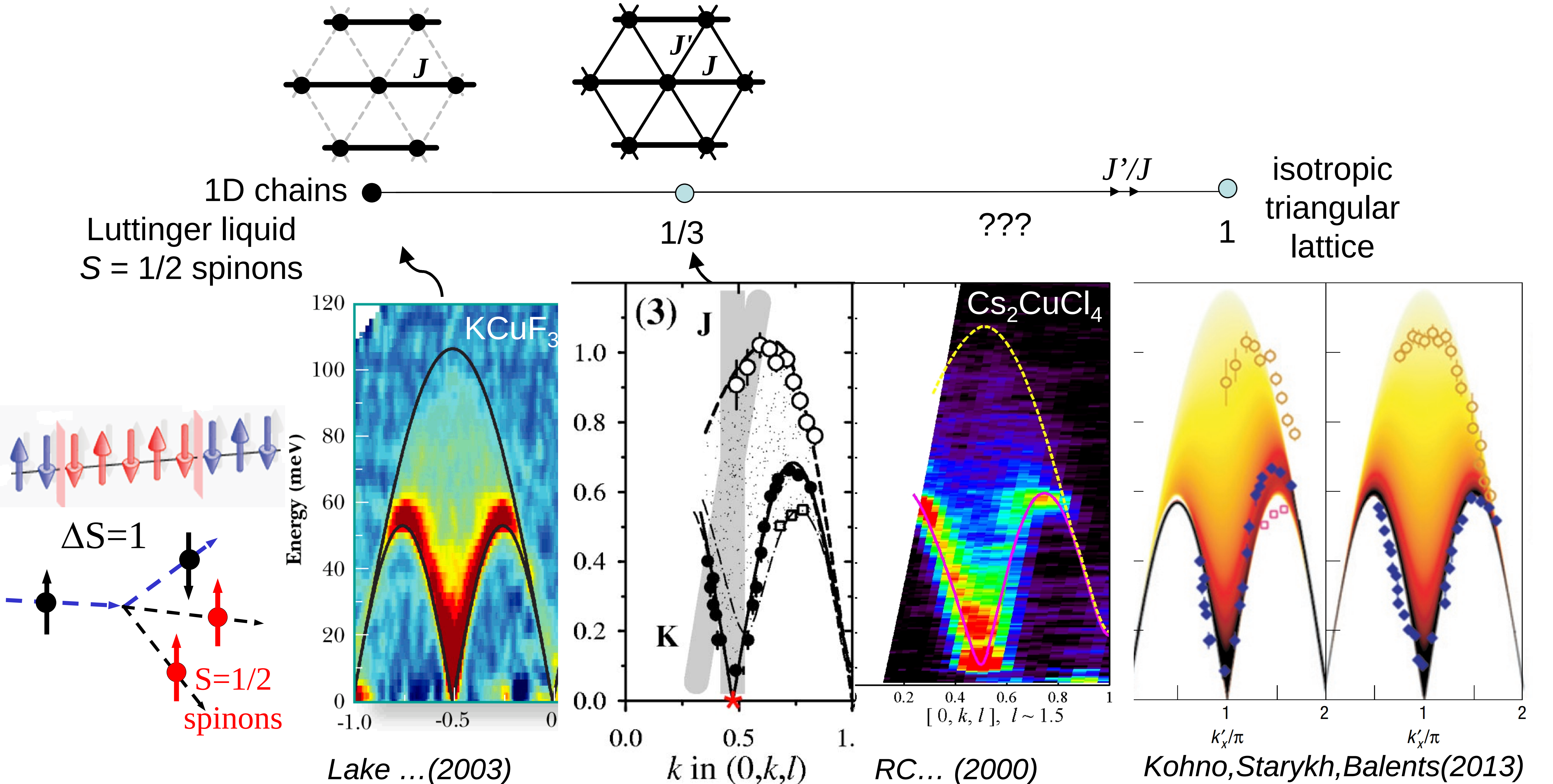




Verresen, Moessner, Pollmann (2019)

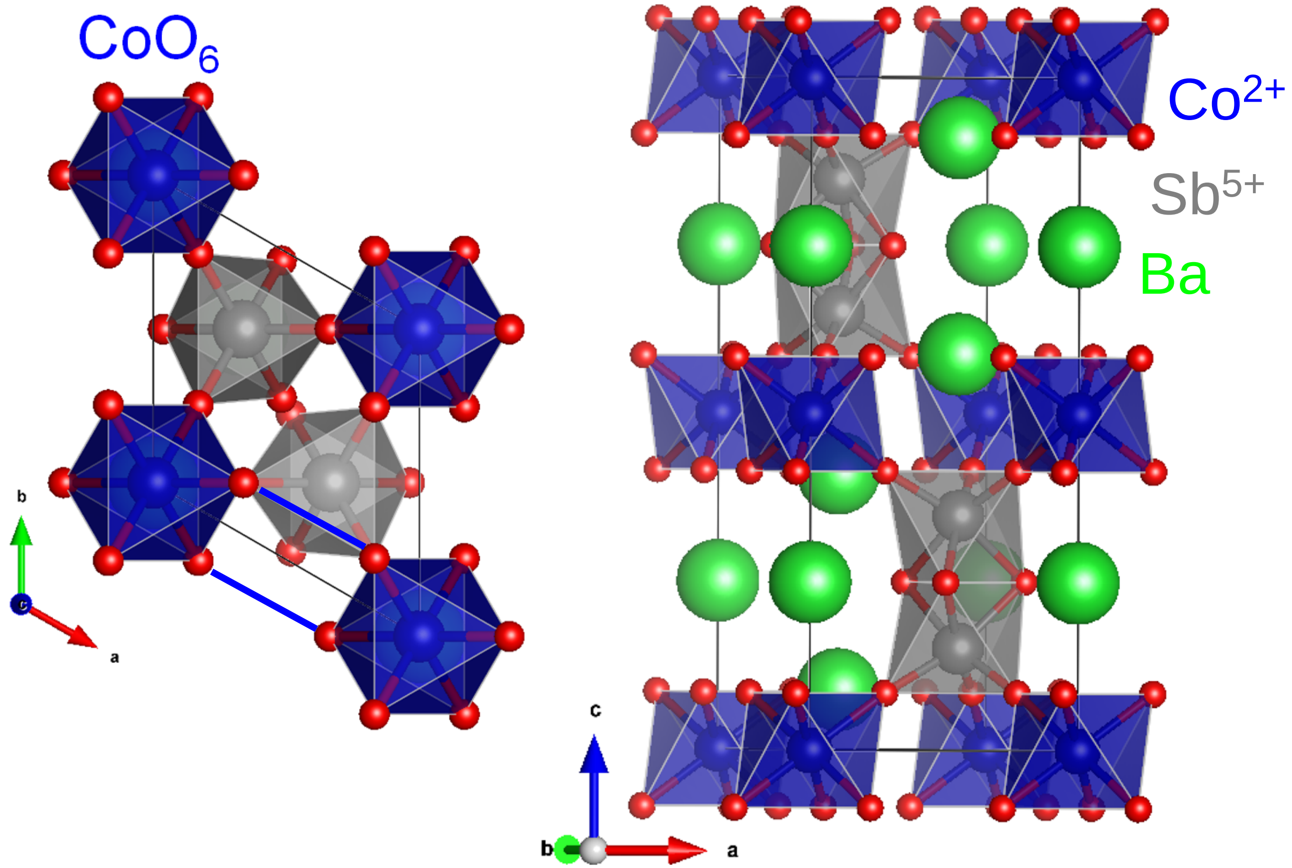



Ferrari, Becca (2019)

# Spatially-anisotropic triangular lattice – phase diagram

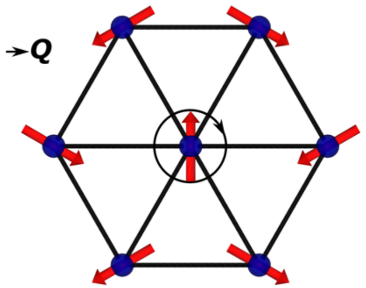


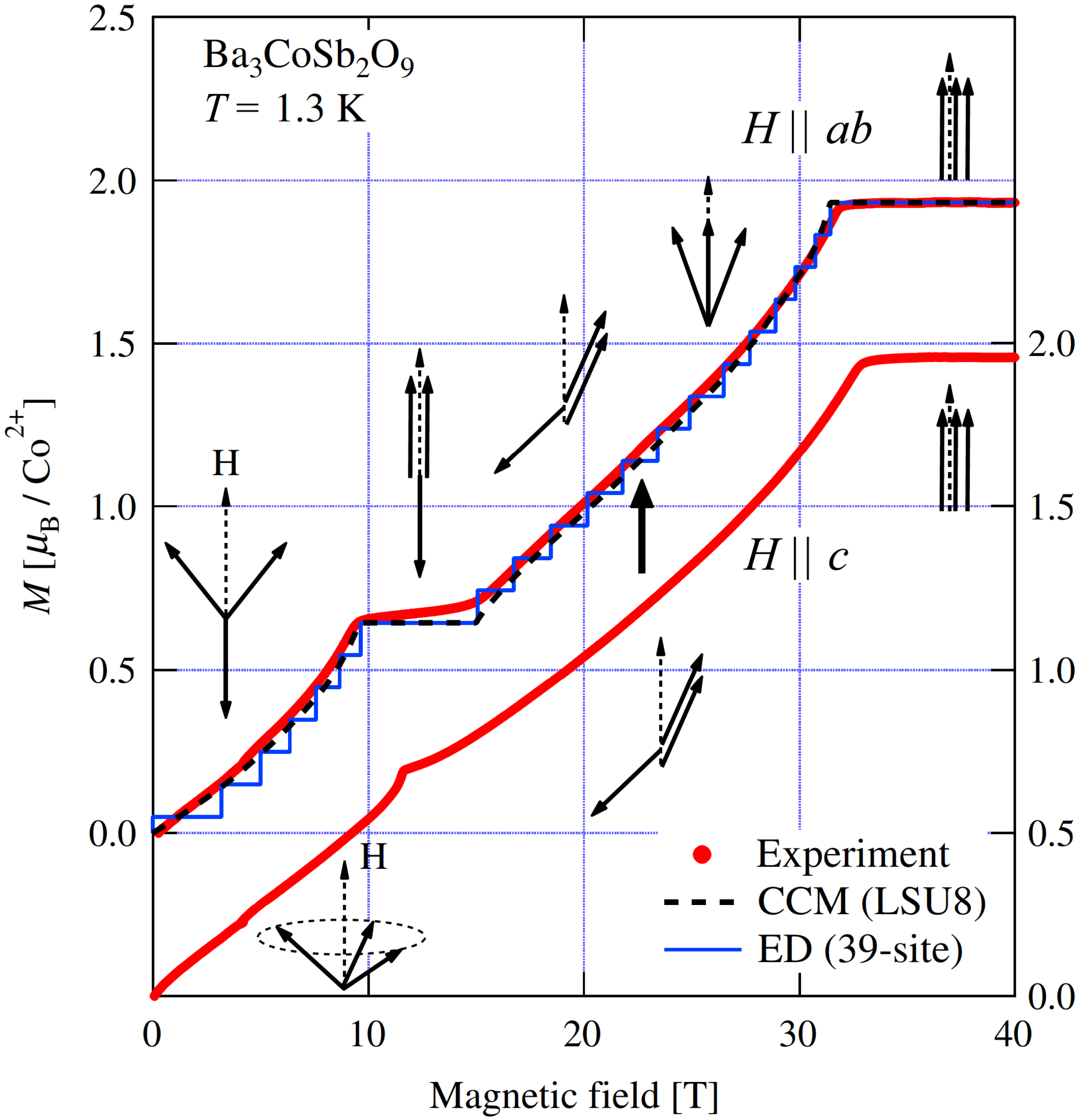
# Hexagonal $Ba_3CoSb_2O_9$




- $CoO_6$ octahedra in triangular layers corner-bonded by $Sb_2O_9$ dumbbells
- 2-layer stacking with 2-fold screw axis (hexagonal *P6$_3$/mmc*) Co $\bar{3}m$ point group

- Co-O-O-Co superexchange AFM
- 120° Nèel order $T_N$~3.7 K
- small XXZ anisotropy
- $Co^{2+}$ Kramers effective spin-1/2

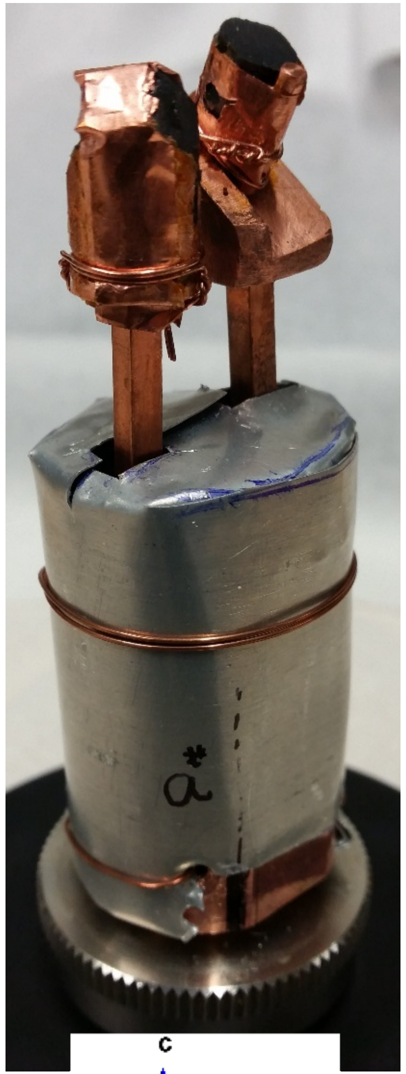


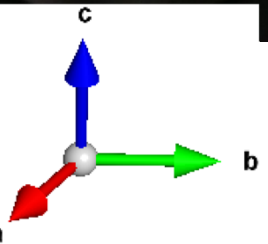




1/3$^{rd}$ magnetization plateau in in-plane field

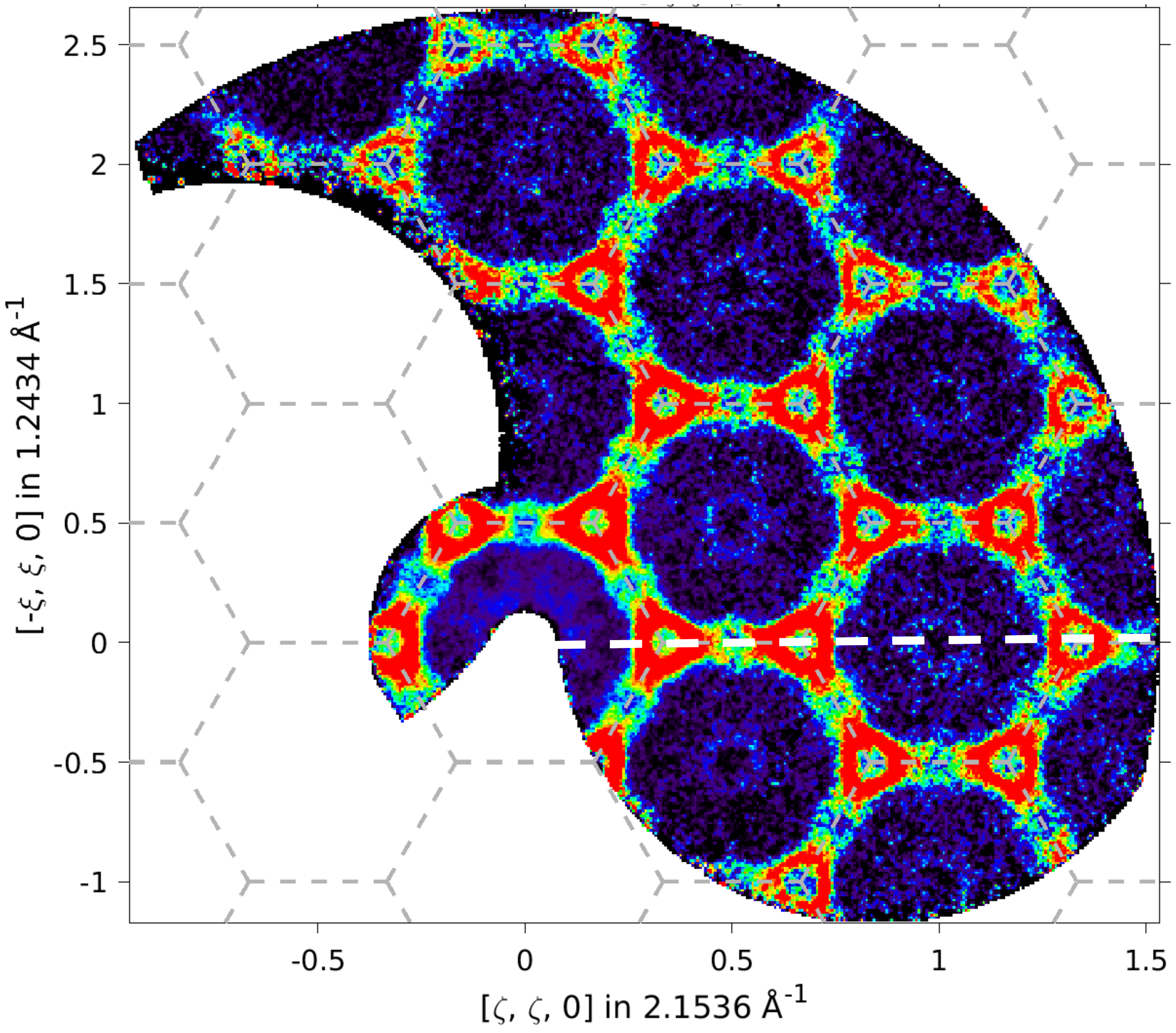
*Suzuki … Tanaka PRL(2013)*

# Inelastic neutron scattering experiments






- mount with (*hk*0) scattering plane horizontal
- cover multiple hexagonal Brillouin zones in-plane
- simultaneously measure multiple incident energies (7 meV – overview, 3.5 meV - higher resolution)



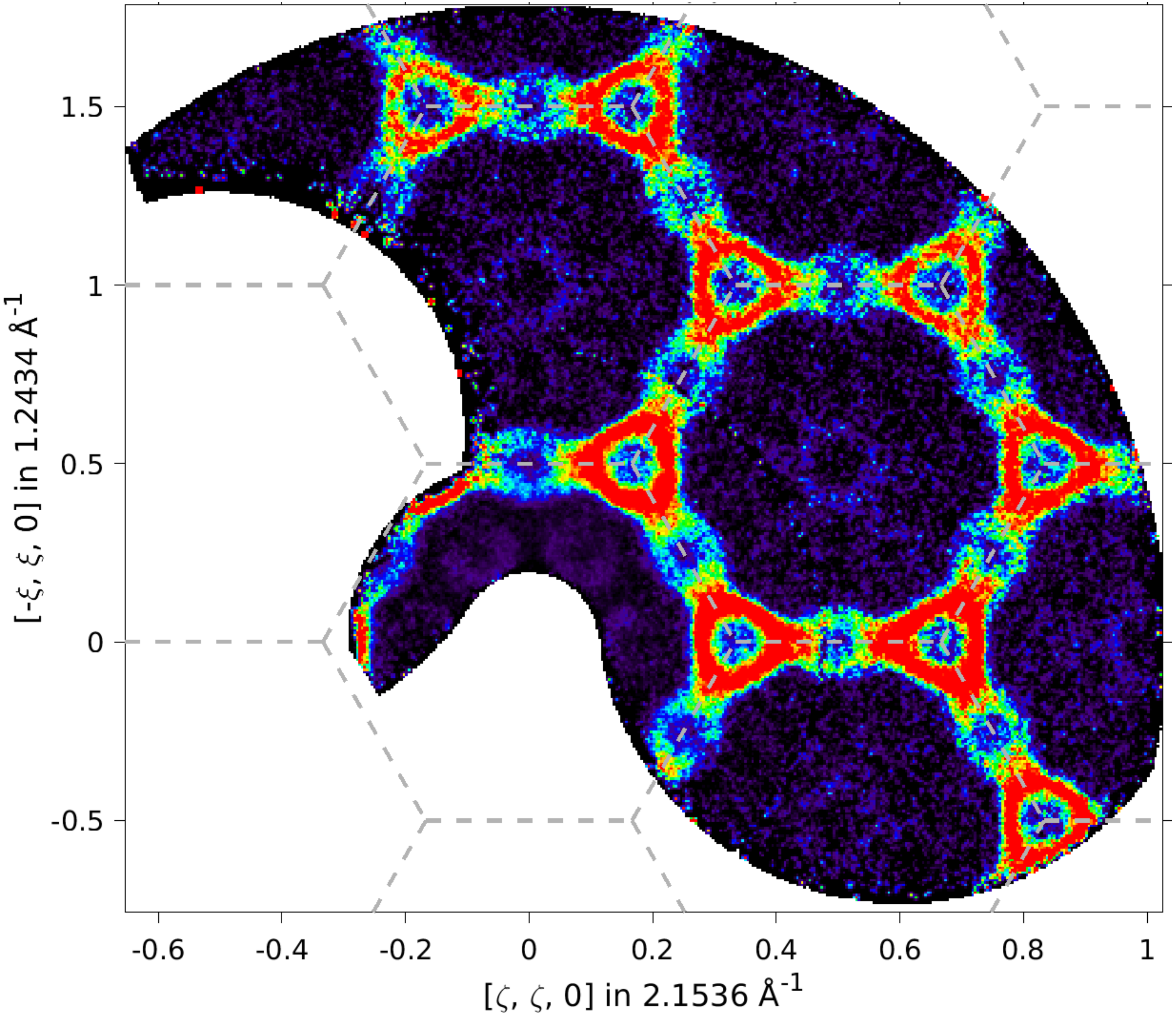

LET @ ISIS

- floating-zone grown crystals of $Ba_3CoSb_2O_9$
- total mass ~ 4 g

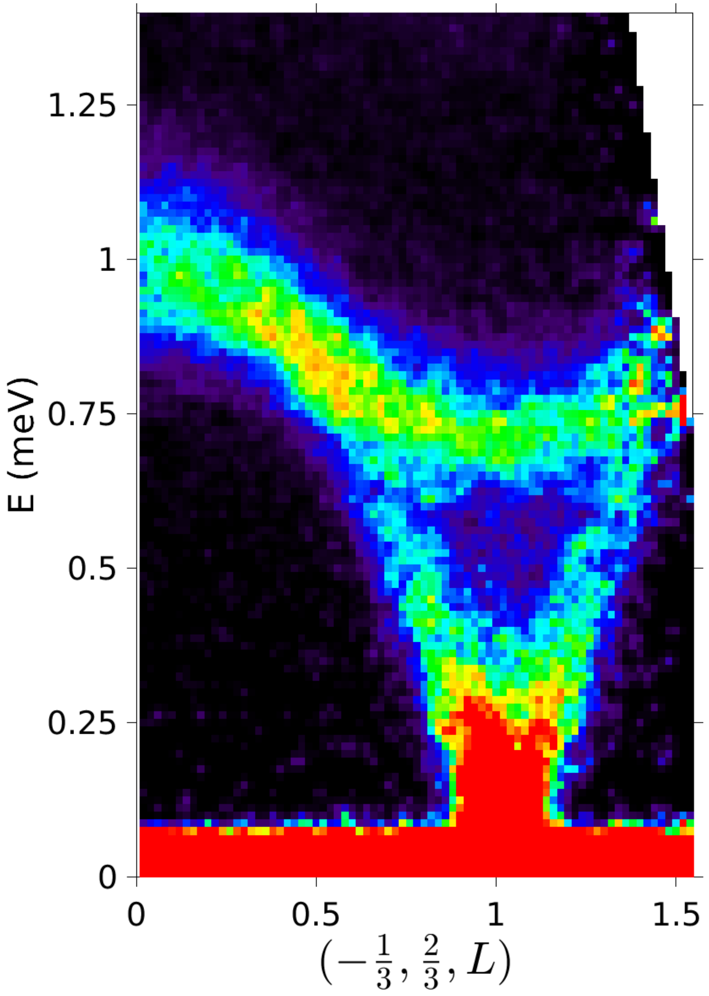
*see also Ito…(2017),Ma... (2016), Zhou…(2012)*

# Inelastic neutron scattering experiments

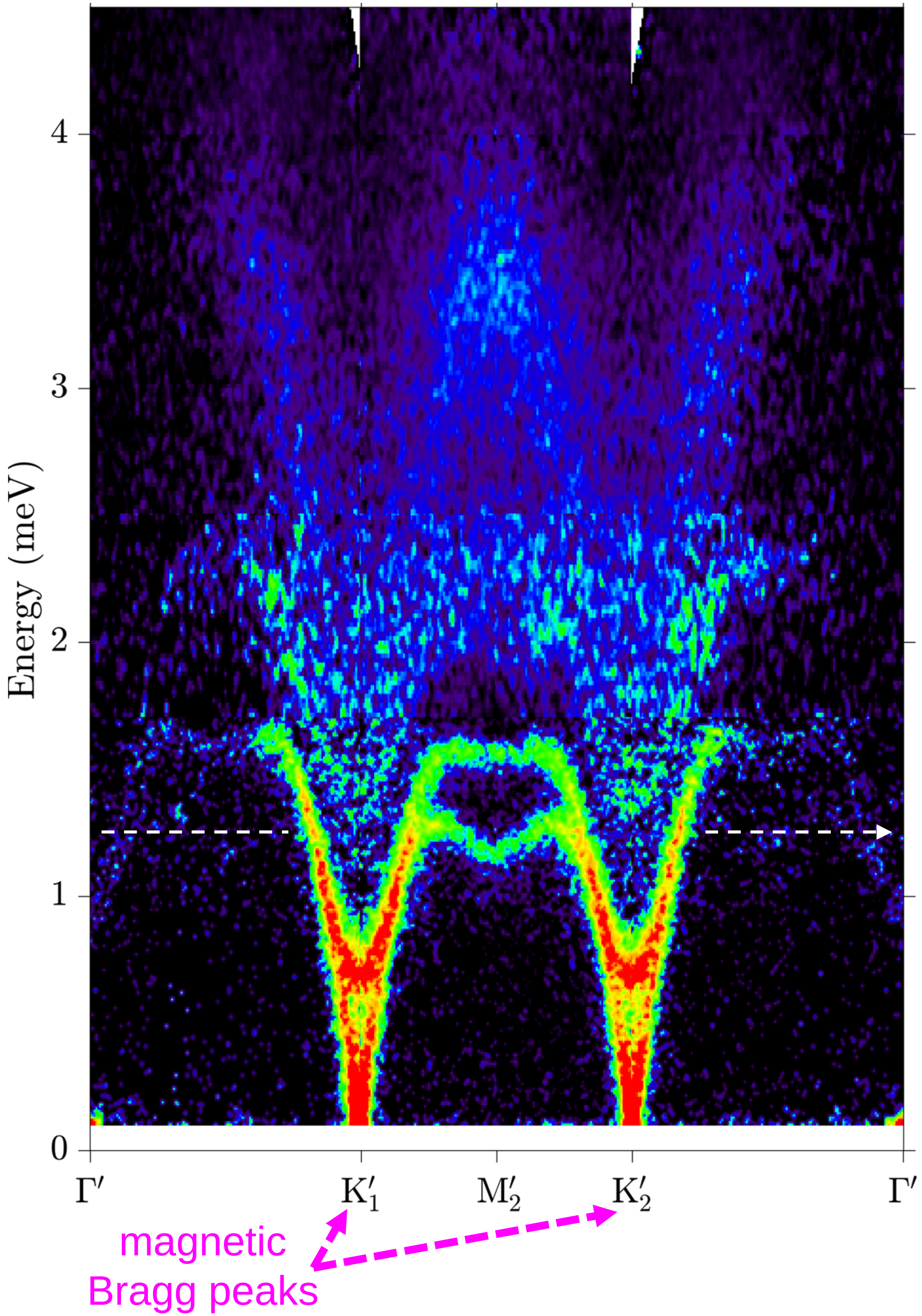
- probe full hexagonal Brillouin zone in-plane with good wavevector resolution



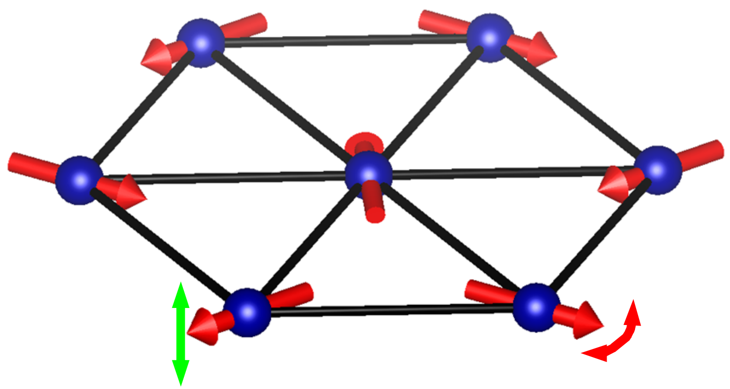

- probe L-dispersion through vertical scattering



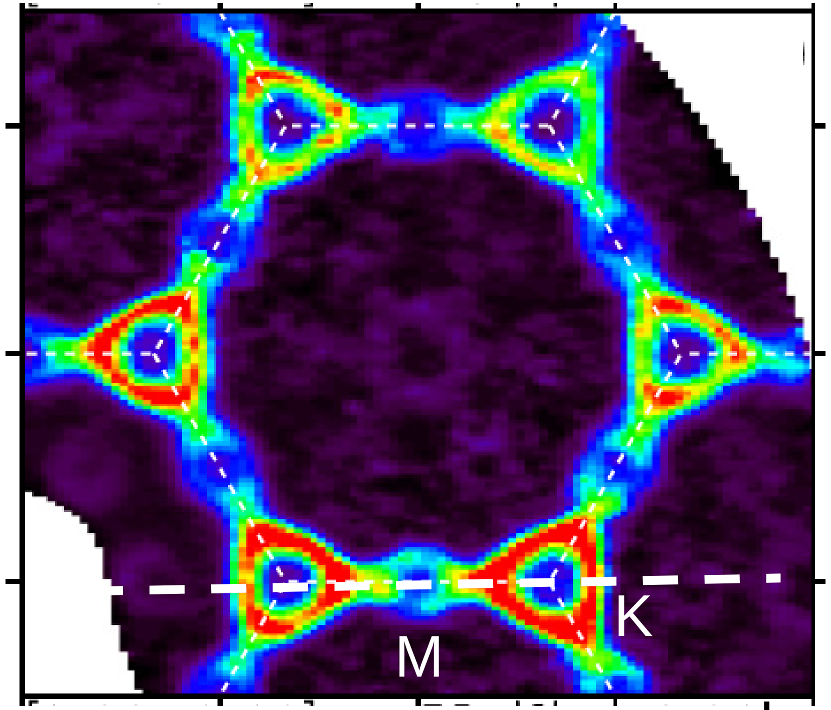

# Overview of the excitation spectrum

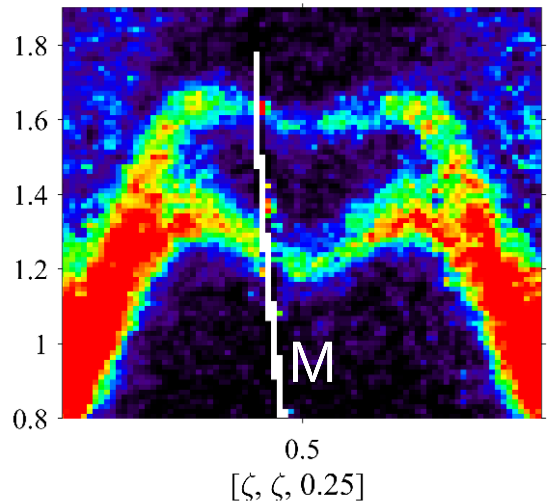


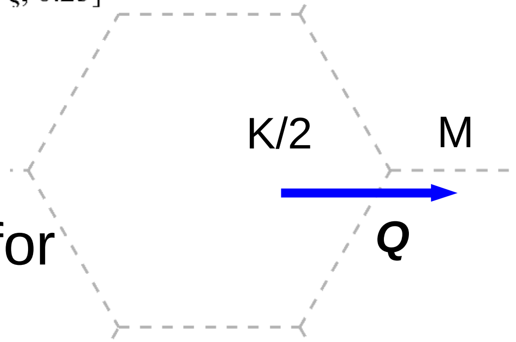

- two intense sharp modes, gapless + gapped above magnetic Bragg peaks characteristic of easy-plane XXZ



- triangular cone at K + oval contours around M due to local "roton" soft mode



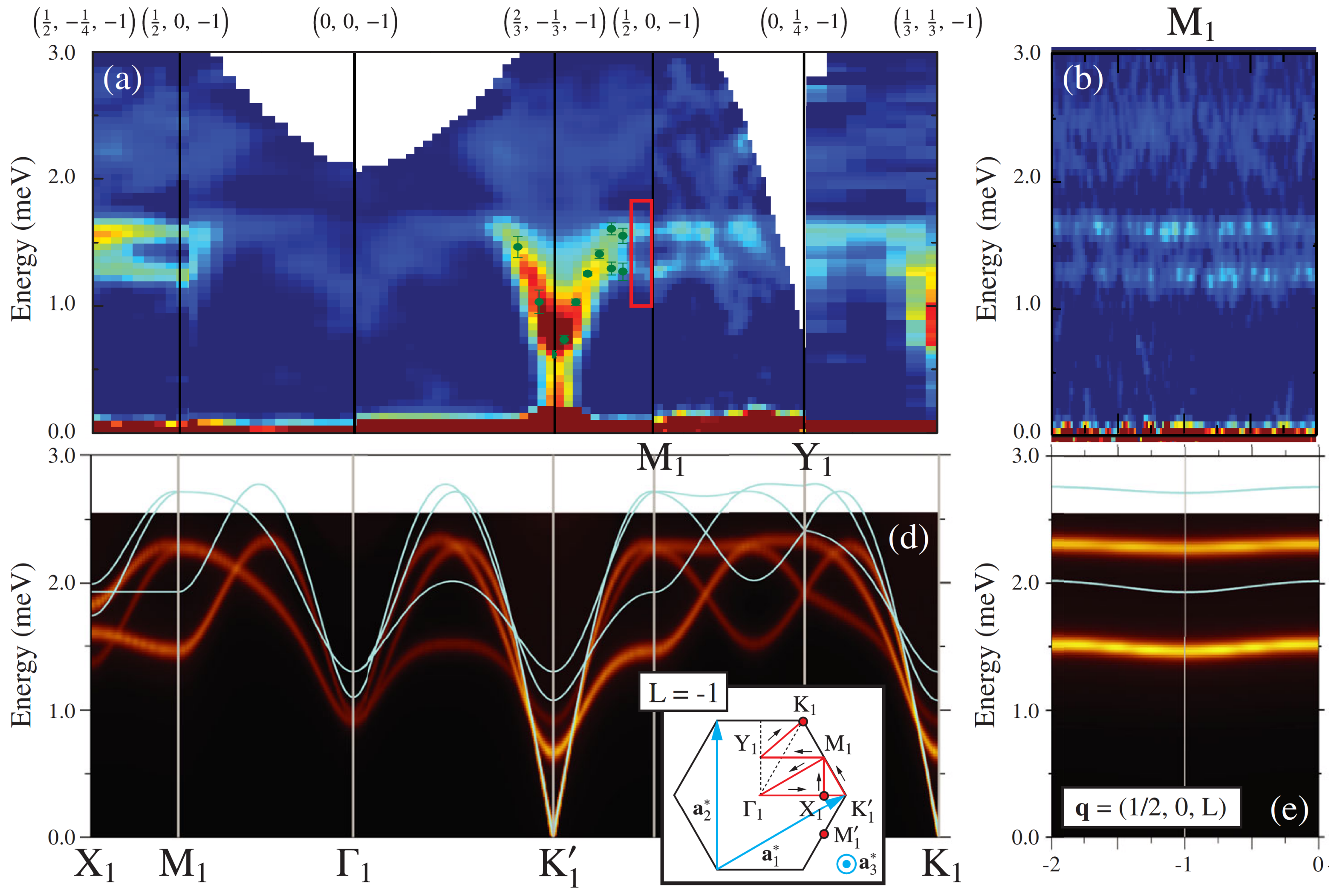




- weaker soft mode in *secondary* mode at M for primary mode ω(K/2)

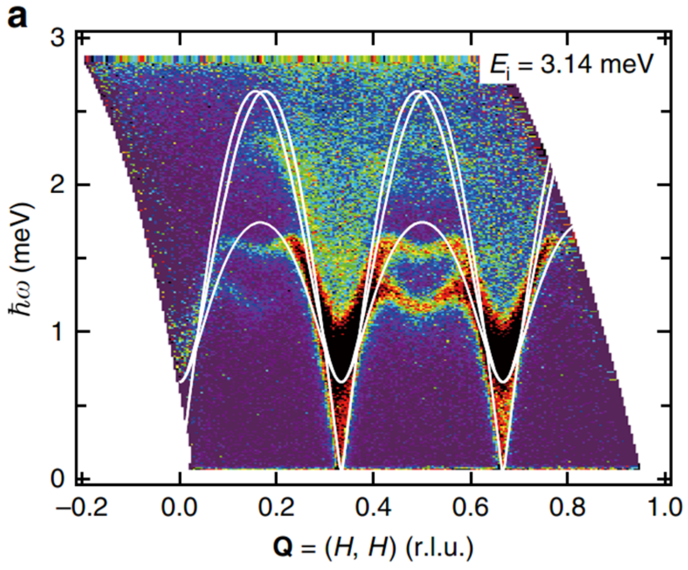
# Dispersions not accounted for by spin wave theory

- allow for $J_1$, interlayer $J_z$, XXZ anisotropy $\Delta$
- capture well low-energy dispersions
- not possible to describe high-energy dispersions even in SWT+1/S (observed $\omega$ ~45% lower)




$$\mathcal{H} = J_1 \sum_{\langle ij \rangle}^{\mathrm{NN}} S_i^x S_j^x + S_i^y S_j^y + \Delta S_i^z S_j^z$$

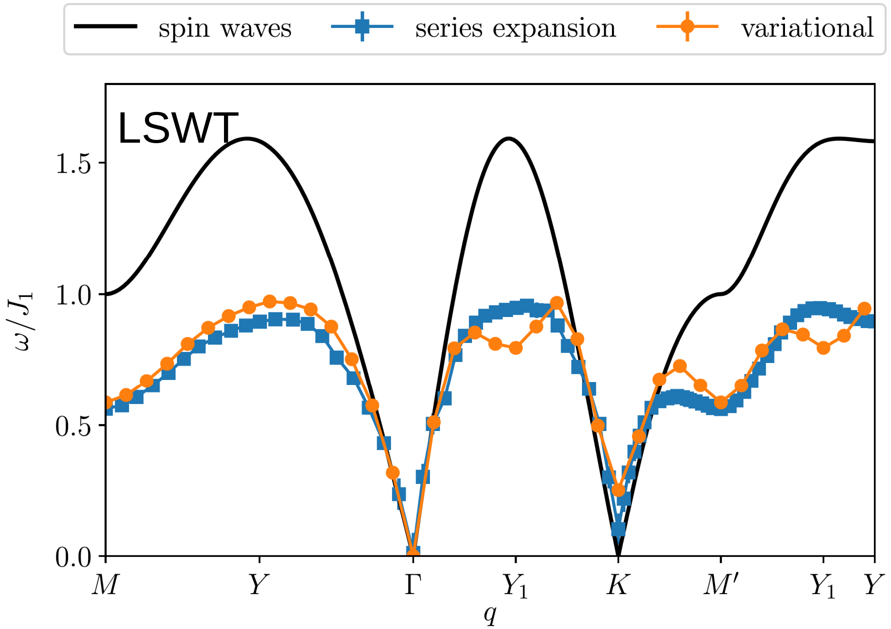
$$+ J_z \sum_{\langle mn \rangle}^{\mathrm{interlayer}} S_m^x S_n^x + S_m^y S_n^y + \Delta S_m^z S_n^z$$

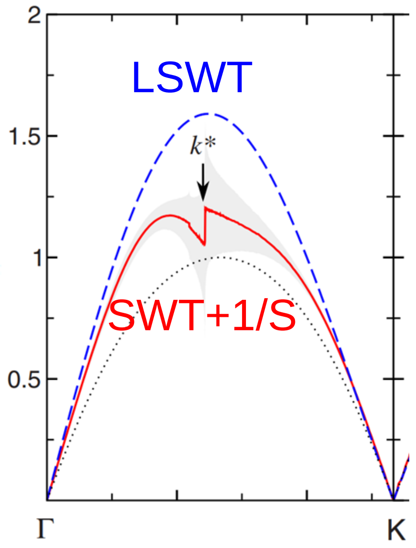


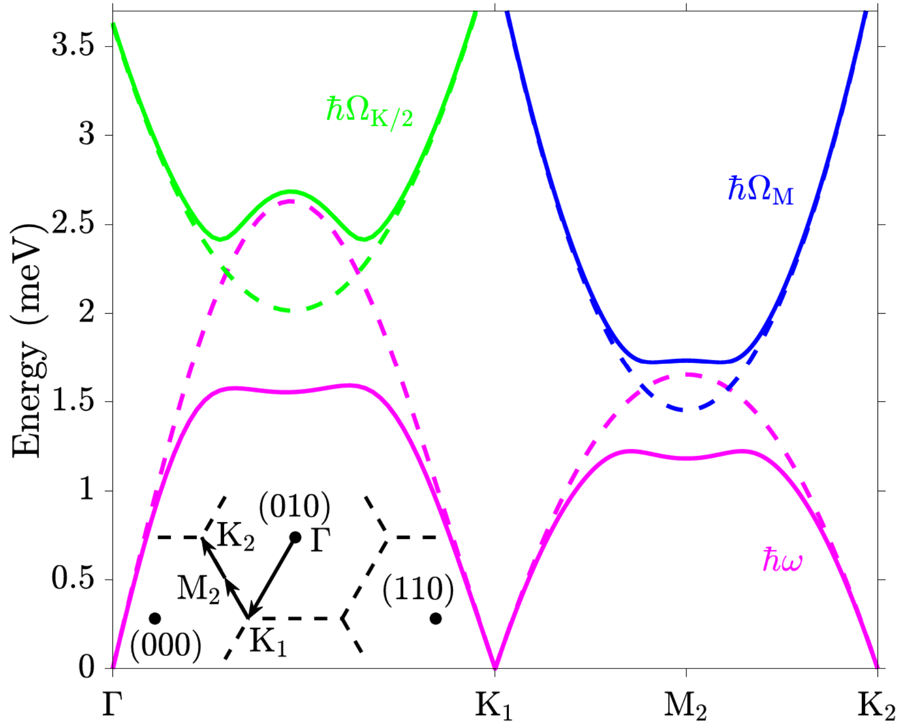

*Ma... (2016)*

*Ito...(2017)*

# Empirical parameterization of the magnon dispersion



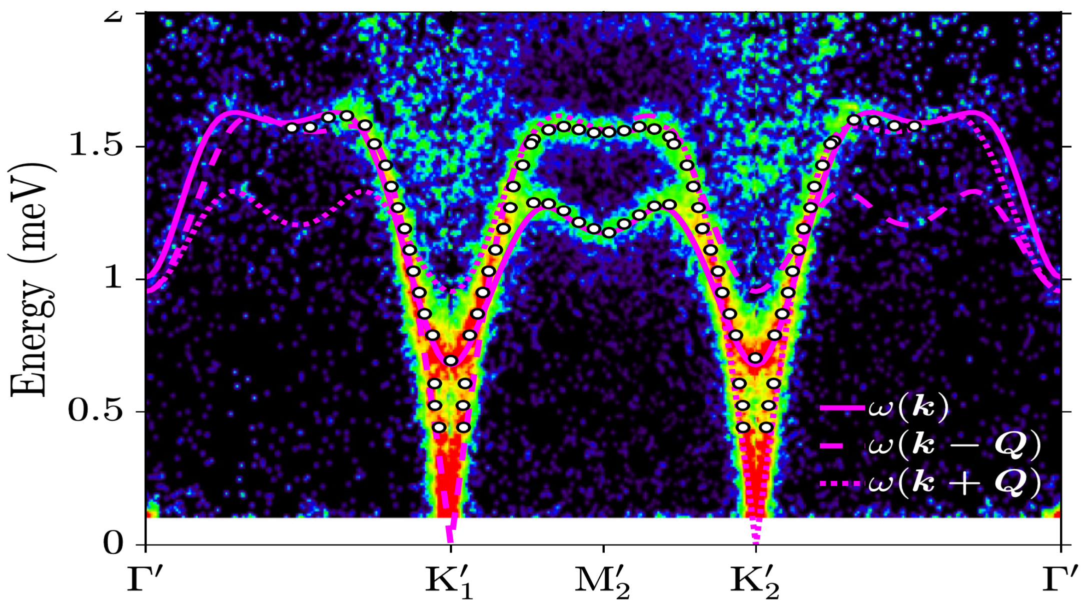



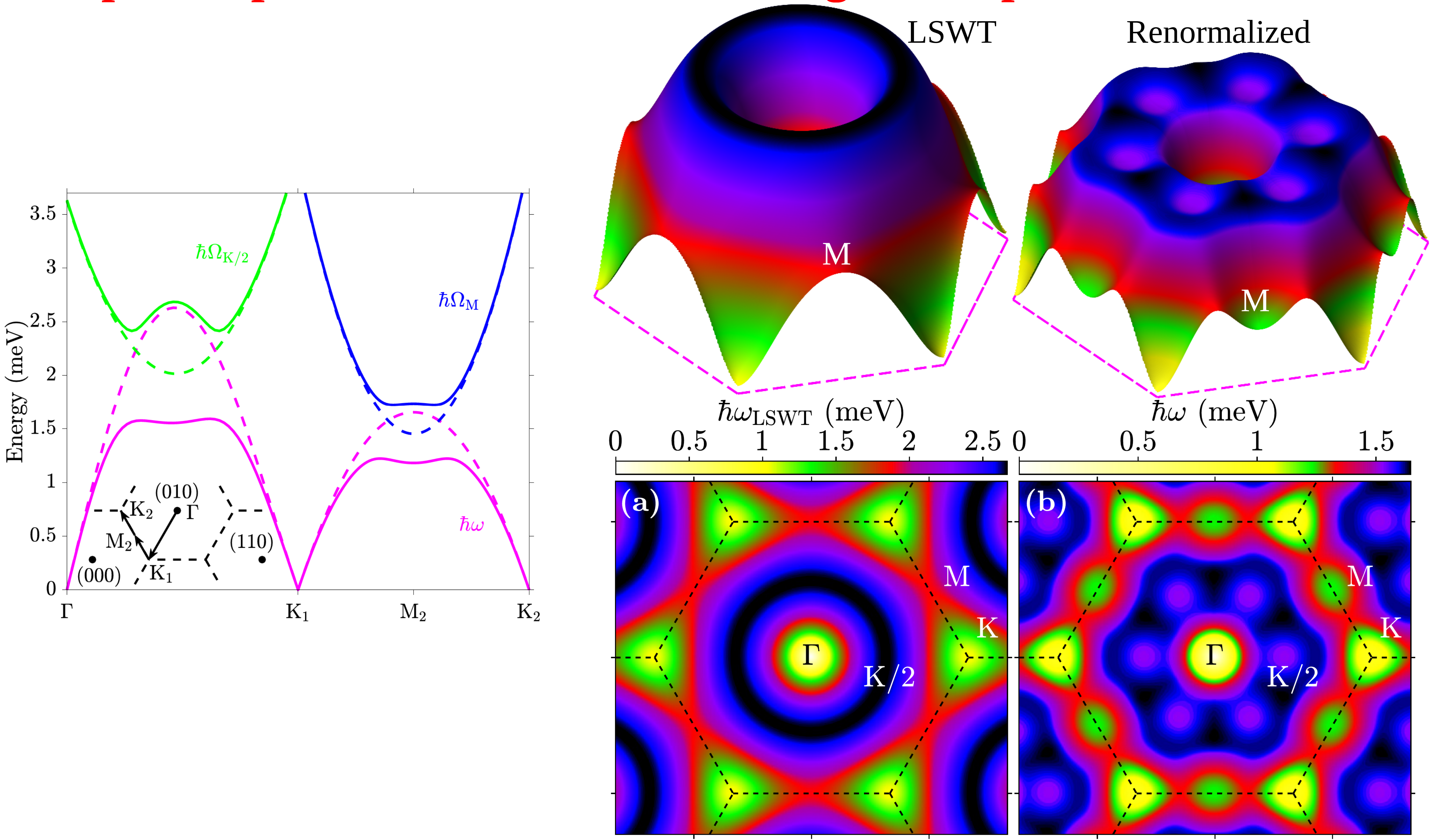

- LSWT expected to work well at low energies
- to capture the soft modes imagine interaction of $\hbar\omega_{\mathrm{LSWT}}$ with a higher energy parabolic mode $\hbar\Omega_i$

$$\begin{pmatrix} \hbar\omega_{\mathrm{LSWT}} & c_i \\ c_i & \hbar\Omega_i \end{pmatrix}$$

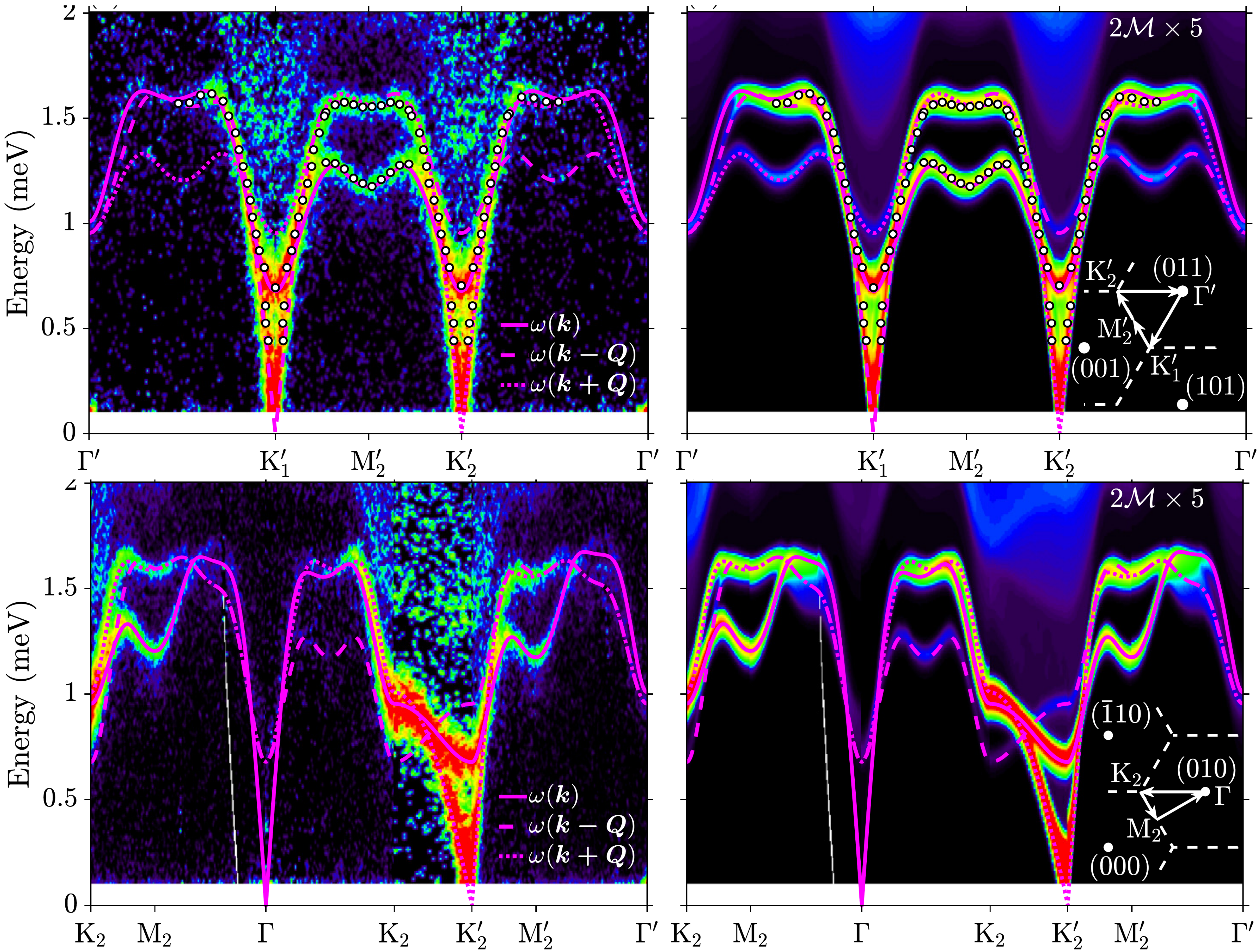
- lower energy eigenvalue $\lambda^-$ inherits the soft mode dip
- motivated empirically to capture repelling effect from interaction with high-energy states, high energy magnons most affected, low energies not affected as expected
- parameterize soft minima at both M and K/2 points



# Empirical parameterization of the magnon dispersion

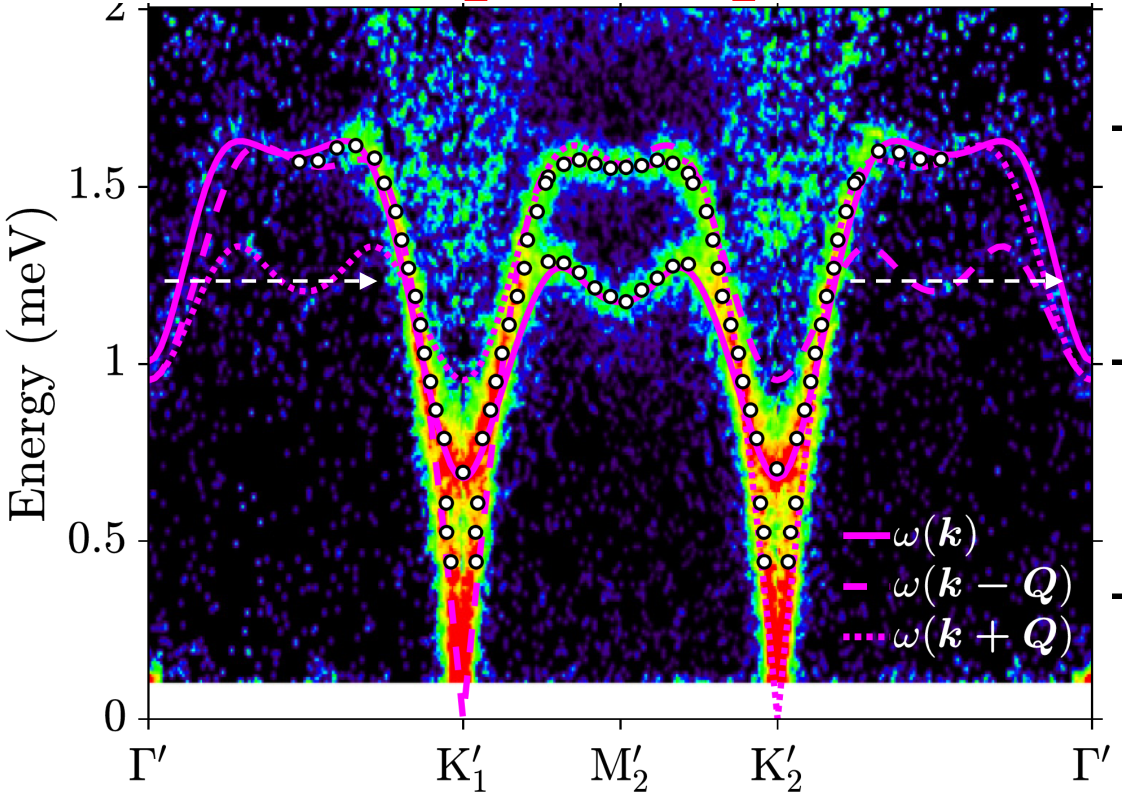


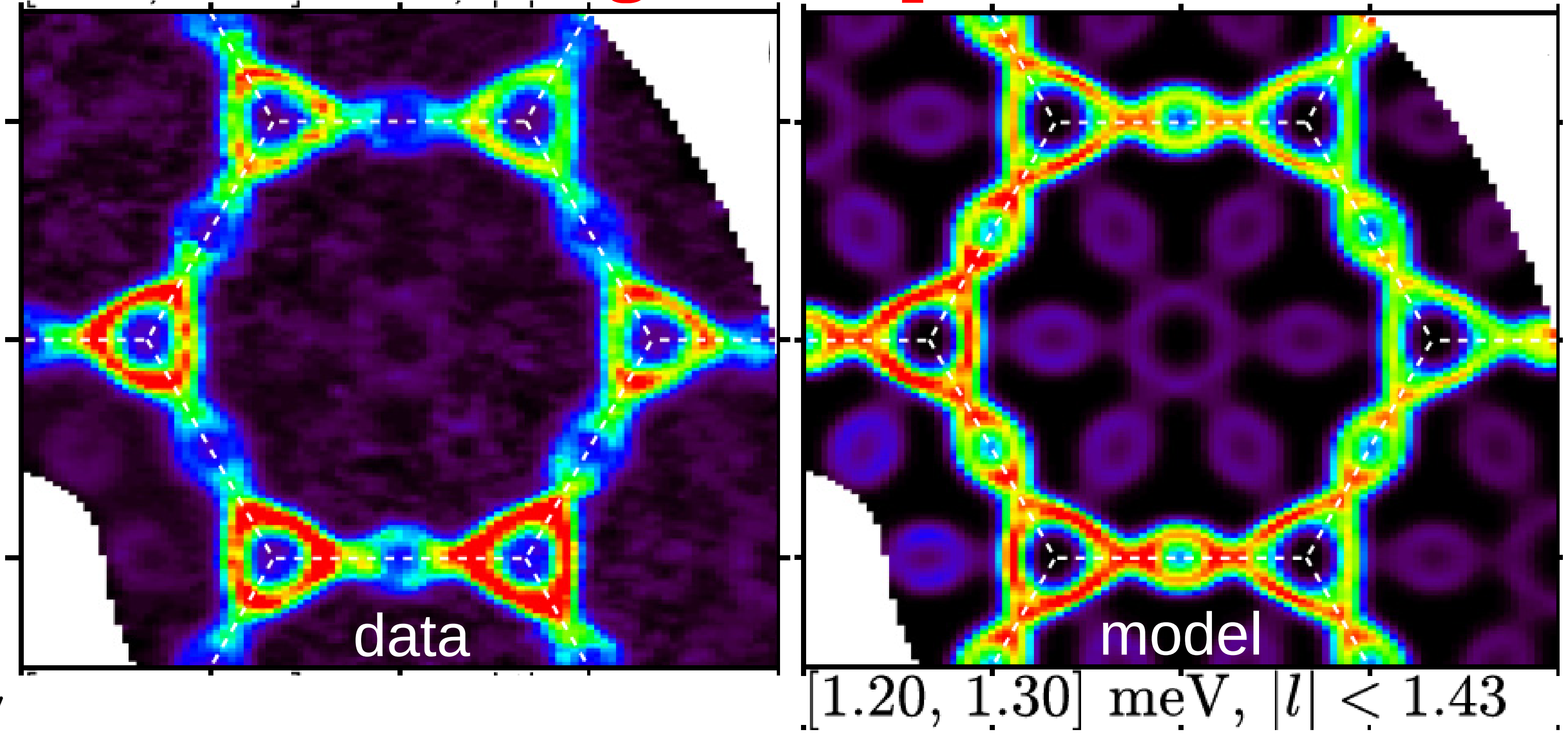
# Empirical parameterization of the magnon dispersion



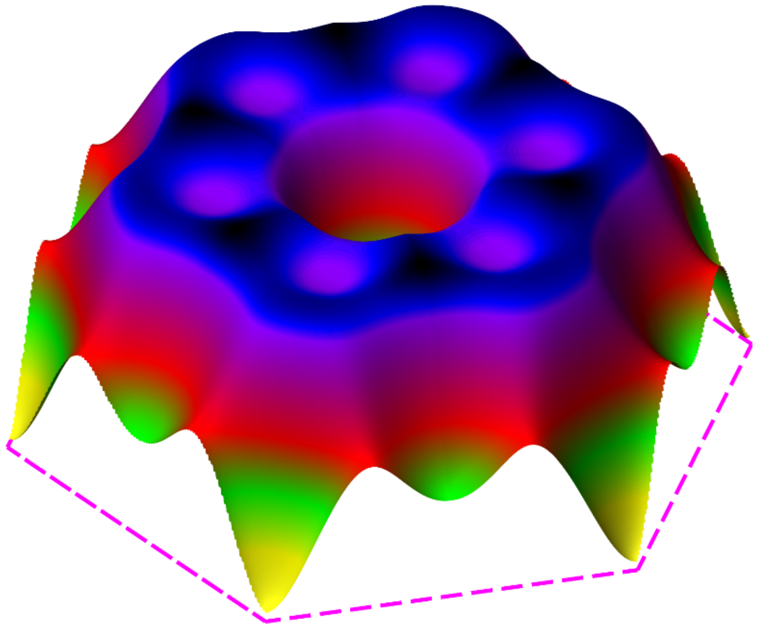

- include nn exchange $J_1$, interlayer coupling $J_z$, XXZ anisotropy $\Delta$
- refine parameterization from global fit to data along many ***k***-directions (include full cross-section model with 3 modes)
- constrain to reproduce saturation field $B_c$
- all dispersions in full 3D Brillouin zone captured quantitatively

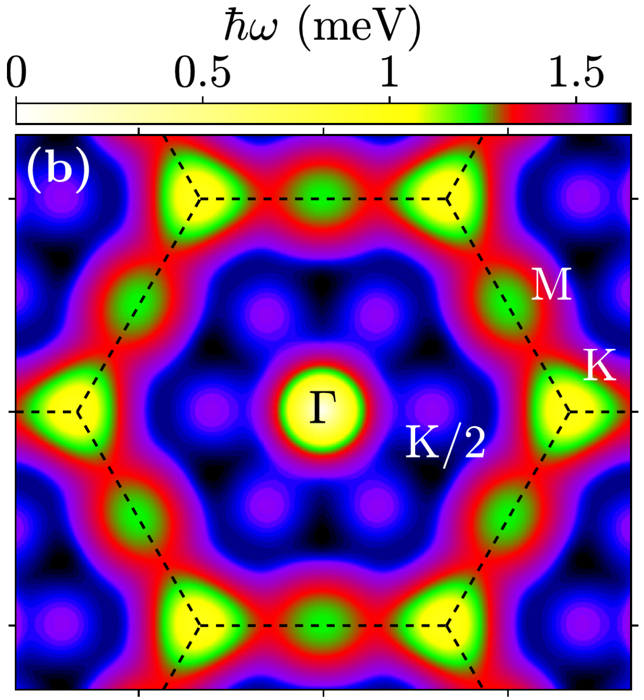
# Empirical parameterization of the magnon dispersion

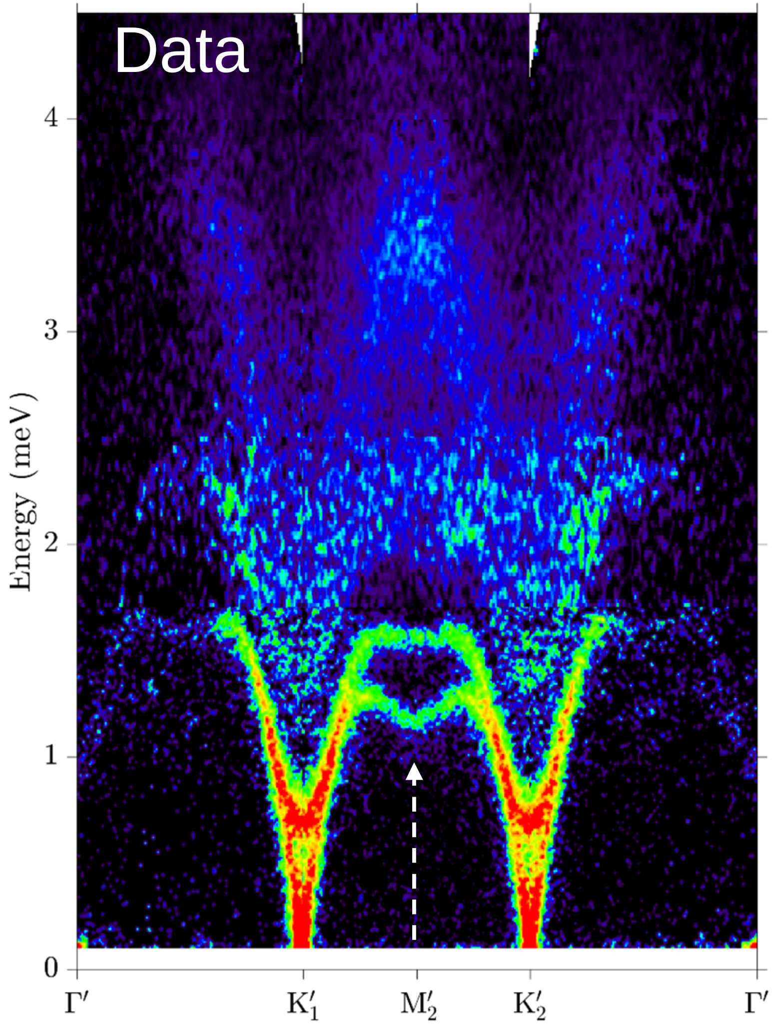


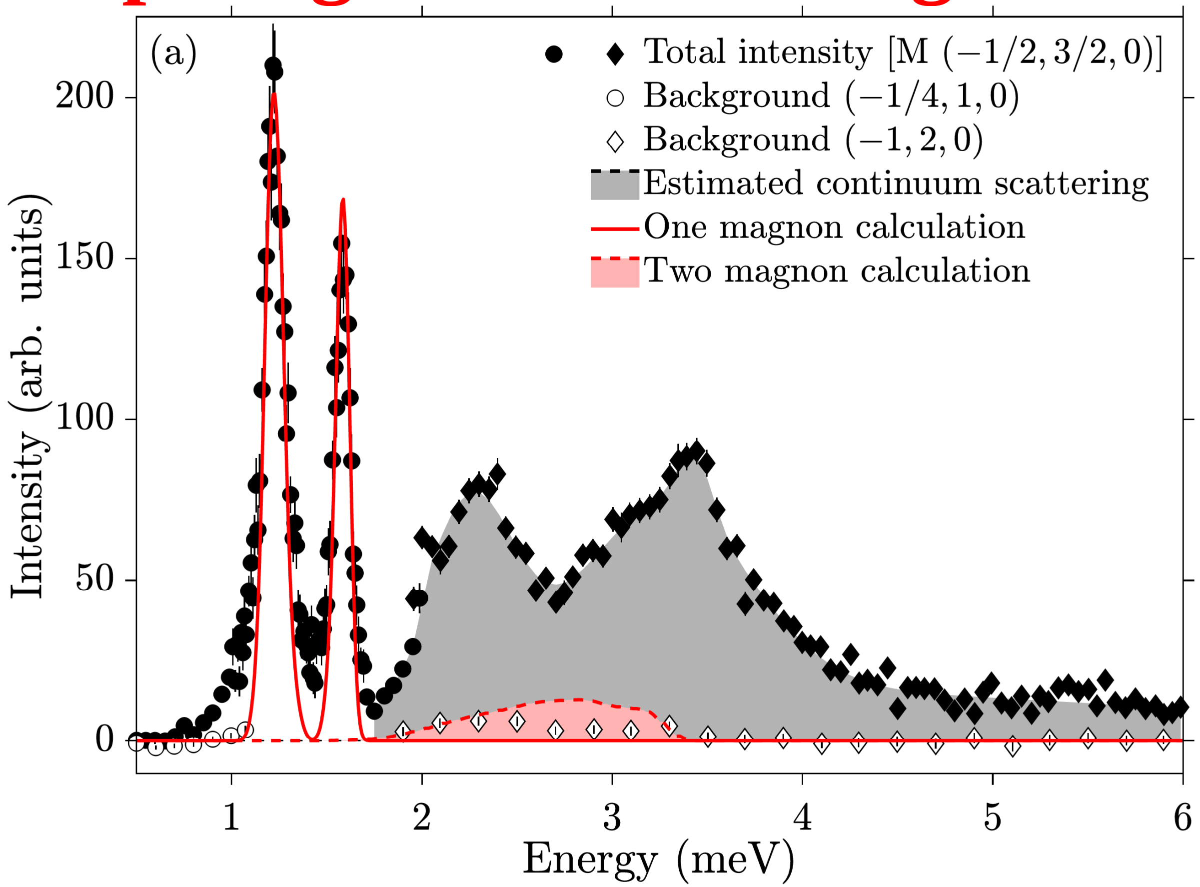

- all dispersion modulations well captured
- soft modes at M and K/2



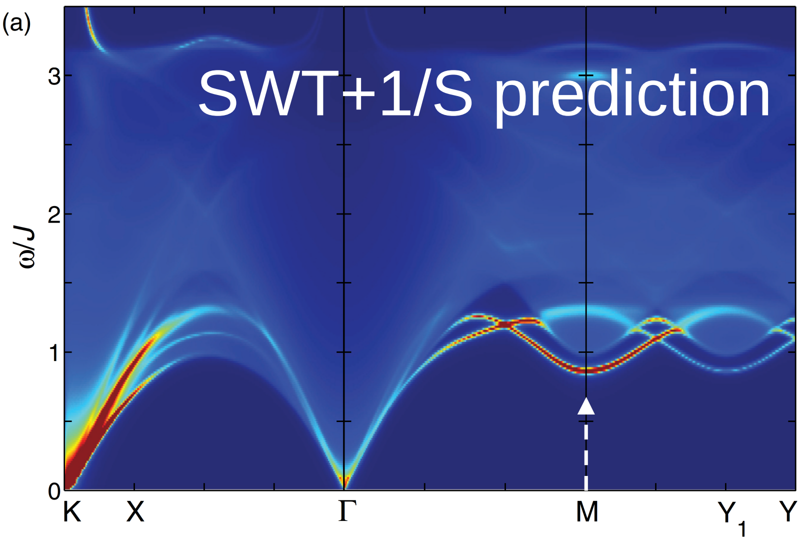


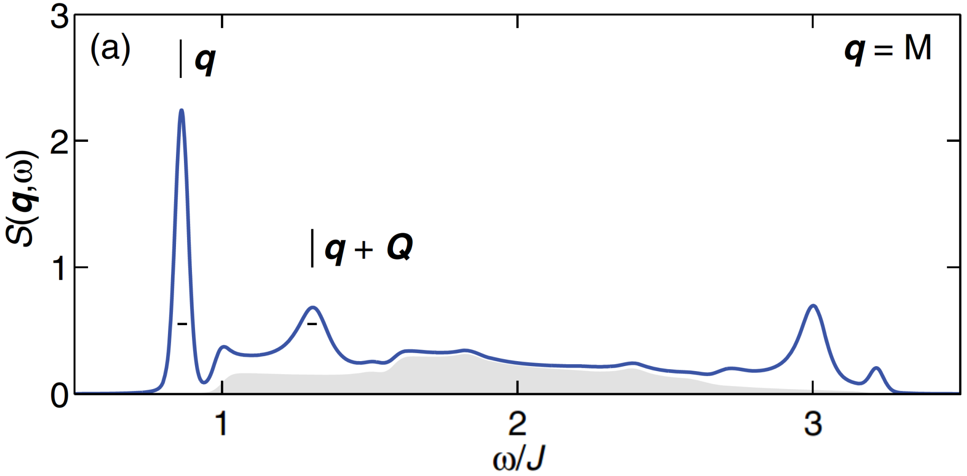



# Sharp magnons and strong continua




- magnons are sharp everywhere, even at top of dispersion



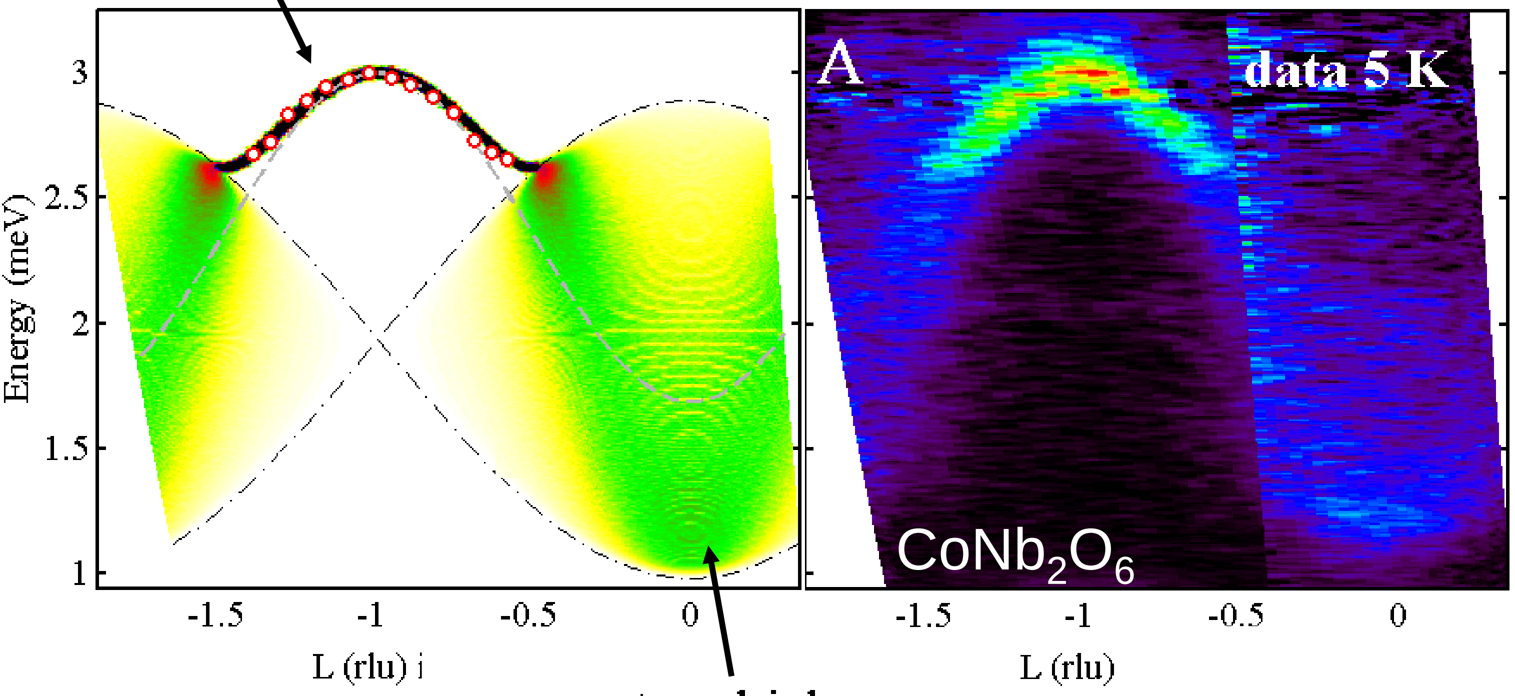



# Decay when a sharp mode enters a continuum

- example of sharp mode entering continuum and decaying

propagating spin flip

↑↑↑•↓•↑↑↑

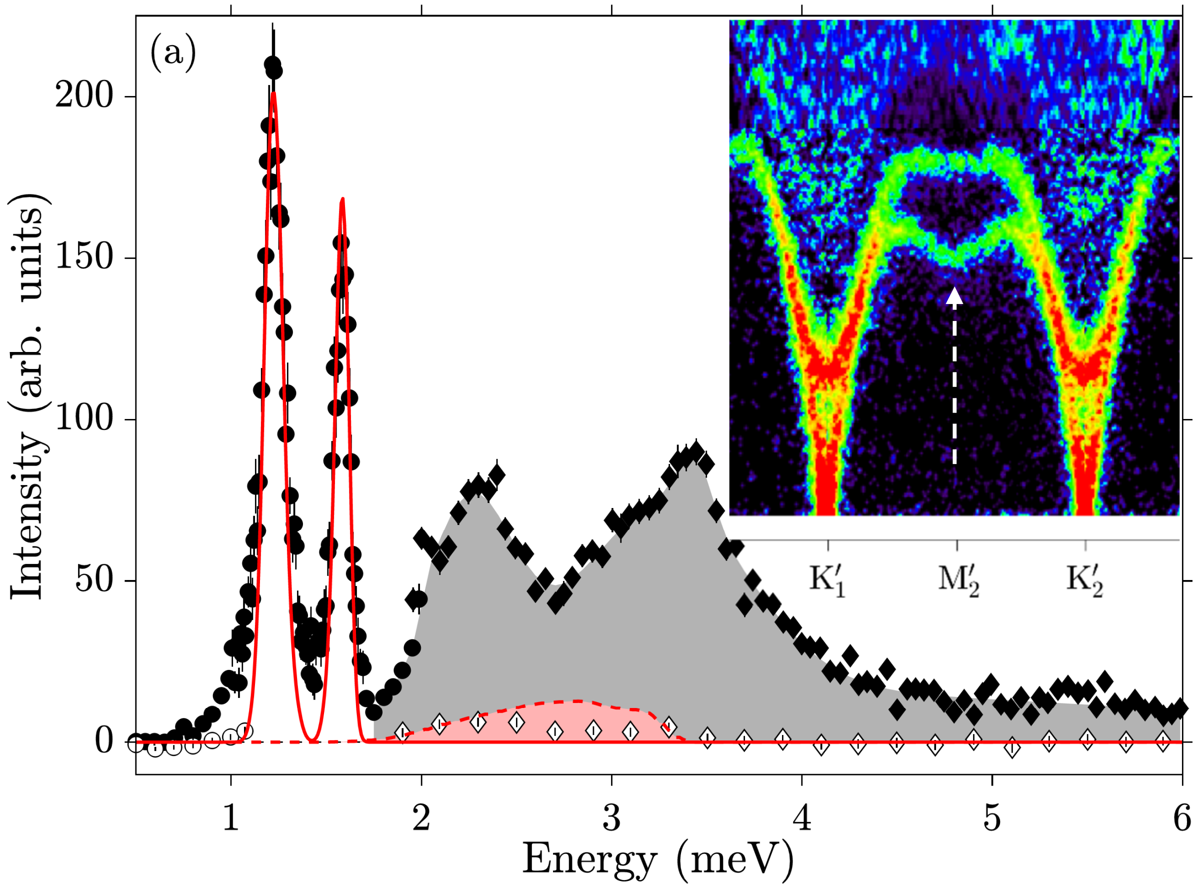



two-kink continuum

↑↑↑↑•↓↓↓•↑↑↑↑

*RC…(2010)*

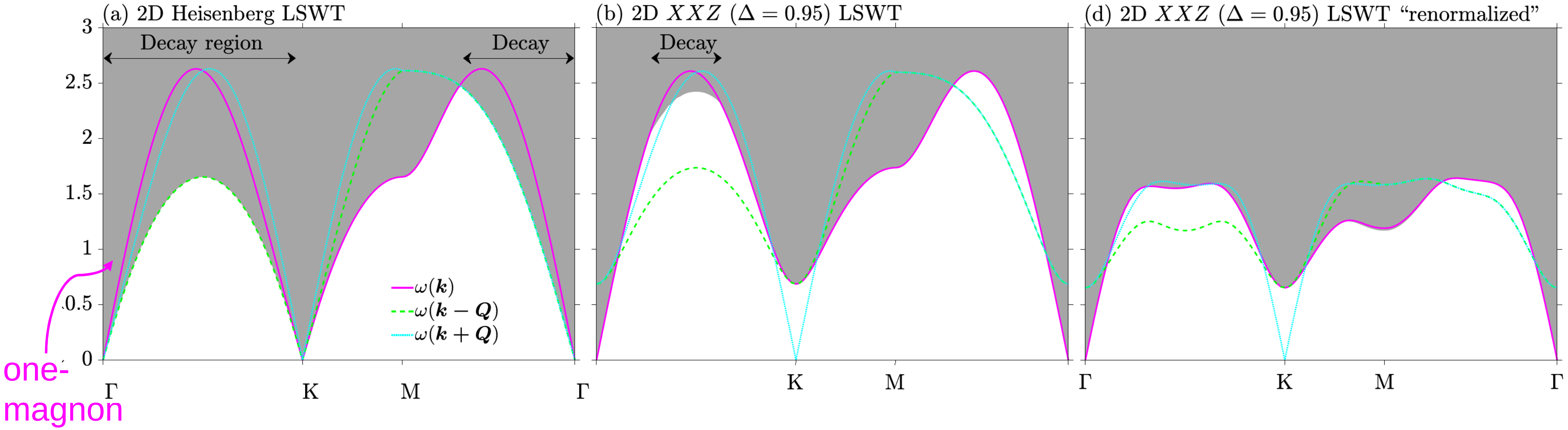
- sharpness of mode suggests it never enters the continuum



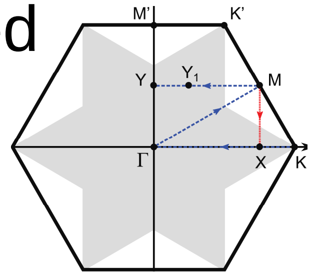
# Why magnons do not decay ?

- two-magnon phase space

$$\boldsymbol{k} = \boldsymbol{k}_1 + \boldsymbol{k}_2$$
$$\omega(\boldsymbol{k}) = \omega(\boldsymbol{k}_1) + \omega(\boldsymbol{k}_2)$$




- Heisenberg LSWT model –> extensive decay region expected

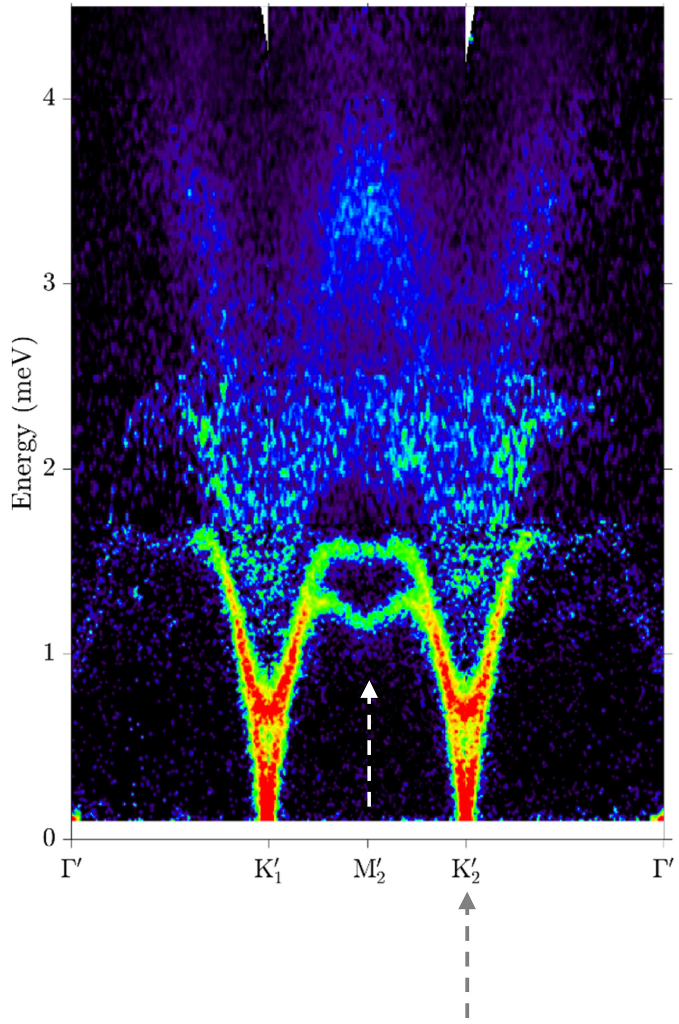



- add anisotropy XXZ –> top magnon still expected to decay

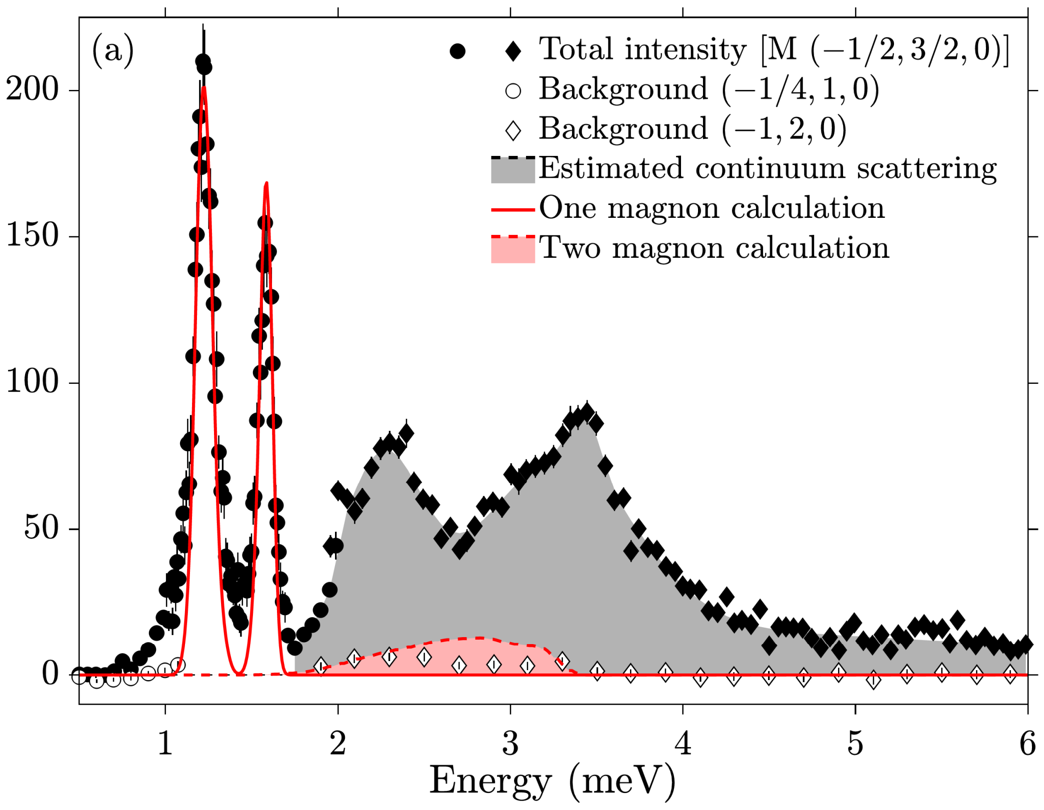
- for the parameterized dispersion no overlap => no decay allowed

# More evidence for quantum interactions: transfer of spectral weight to continuum

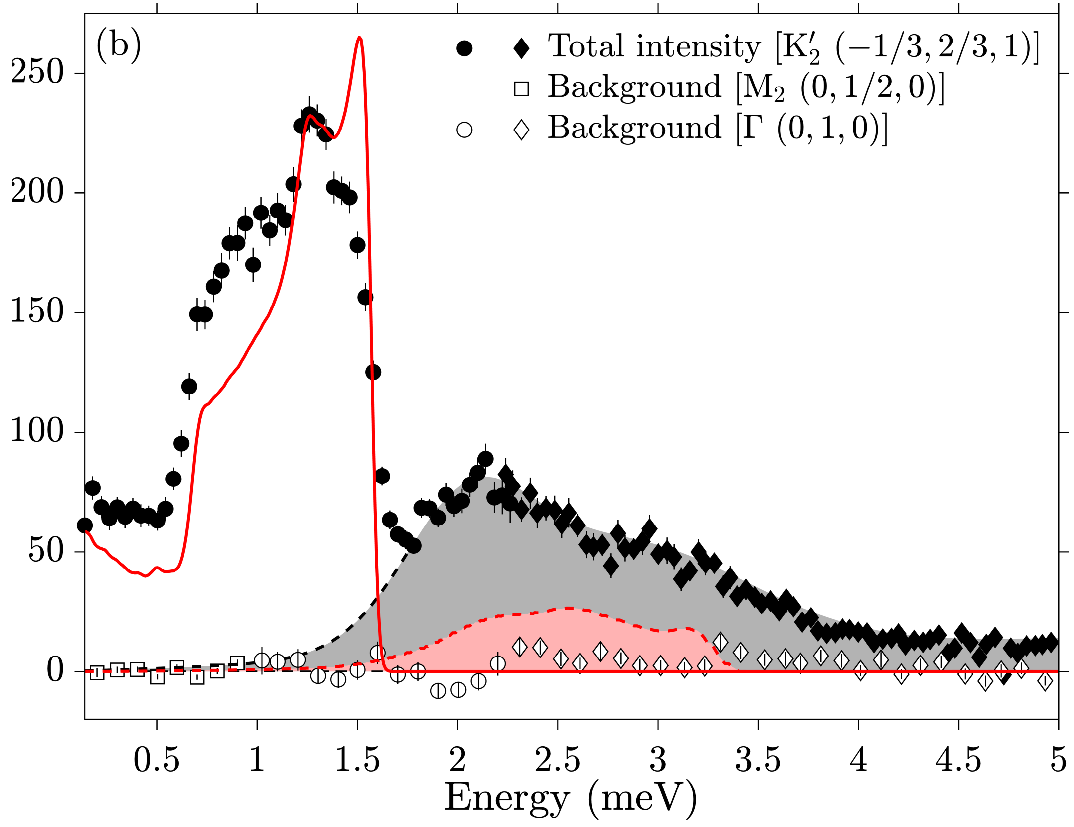
- overall weight of continuum scattering much stronger than expected two-magnon (LSWT)
- magnon intensity decreases faster with increasing ω than 1/ω expected by LSWT
- high-energy magnons transfer part of their weight to continuum




scan at M

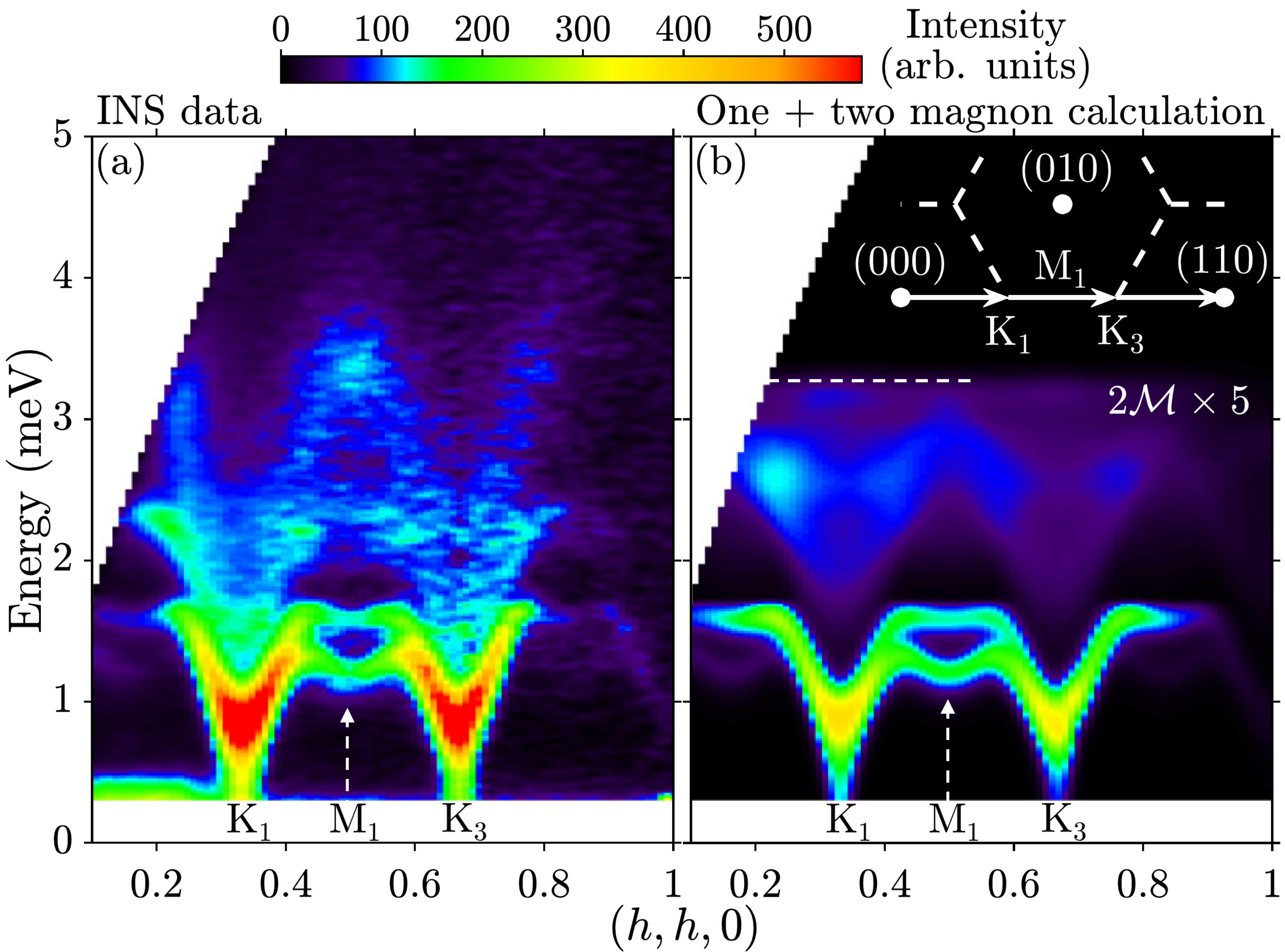


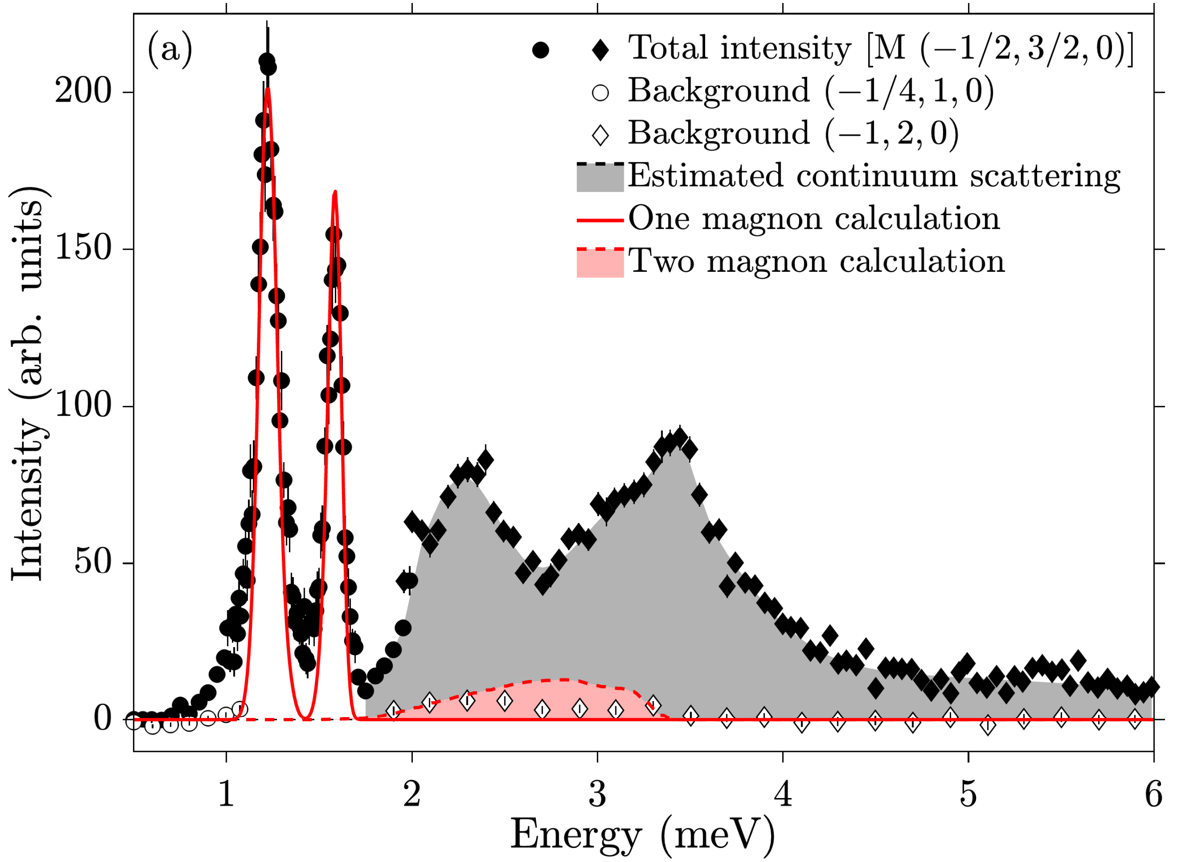

scan at $K_2$’



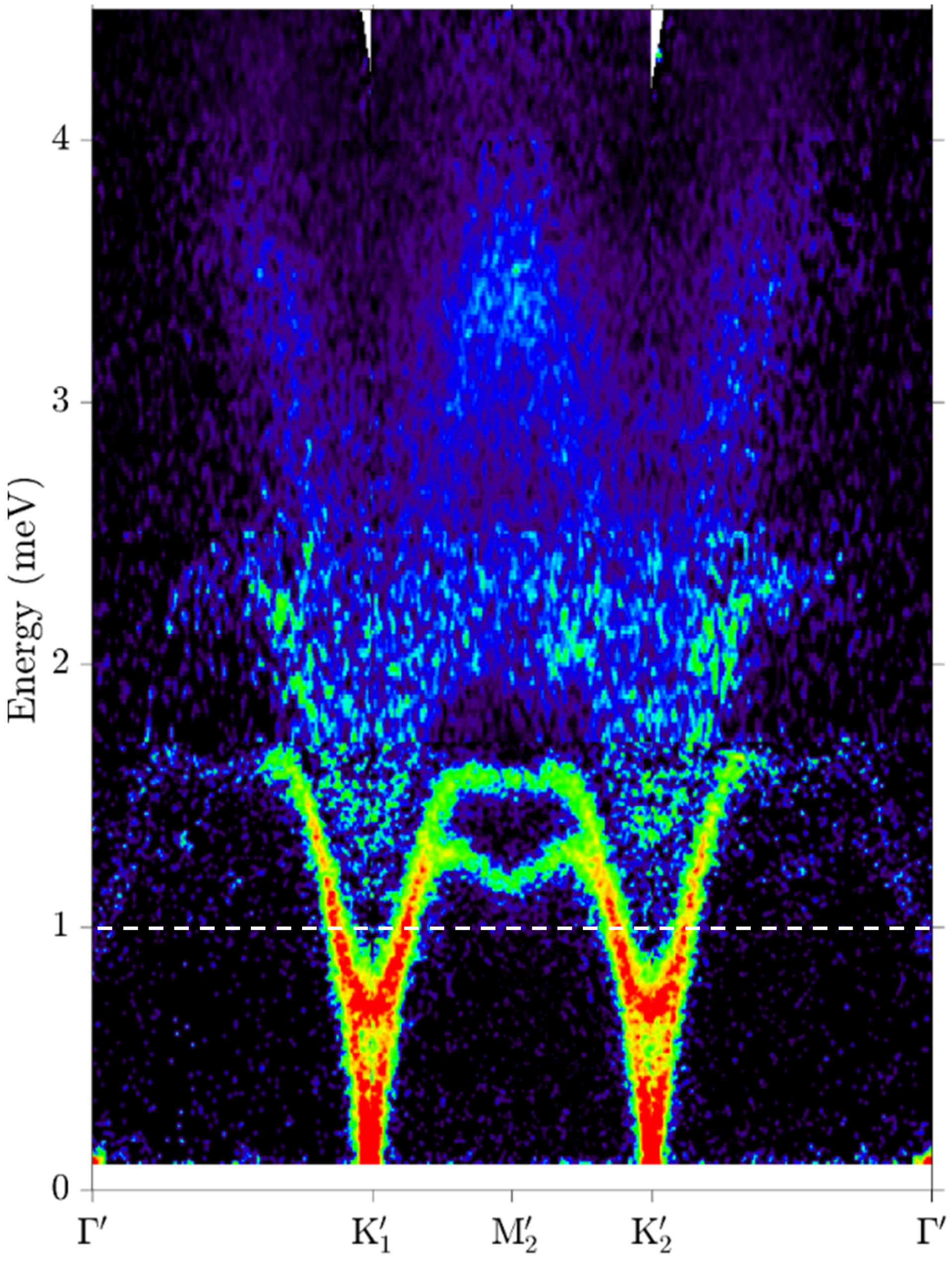
# Continuum scattering compared to a two-magnon cross-section

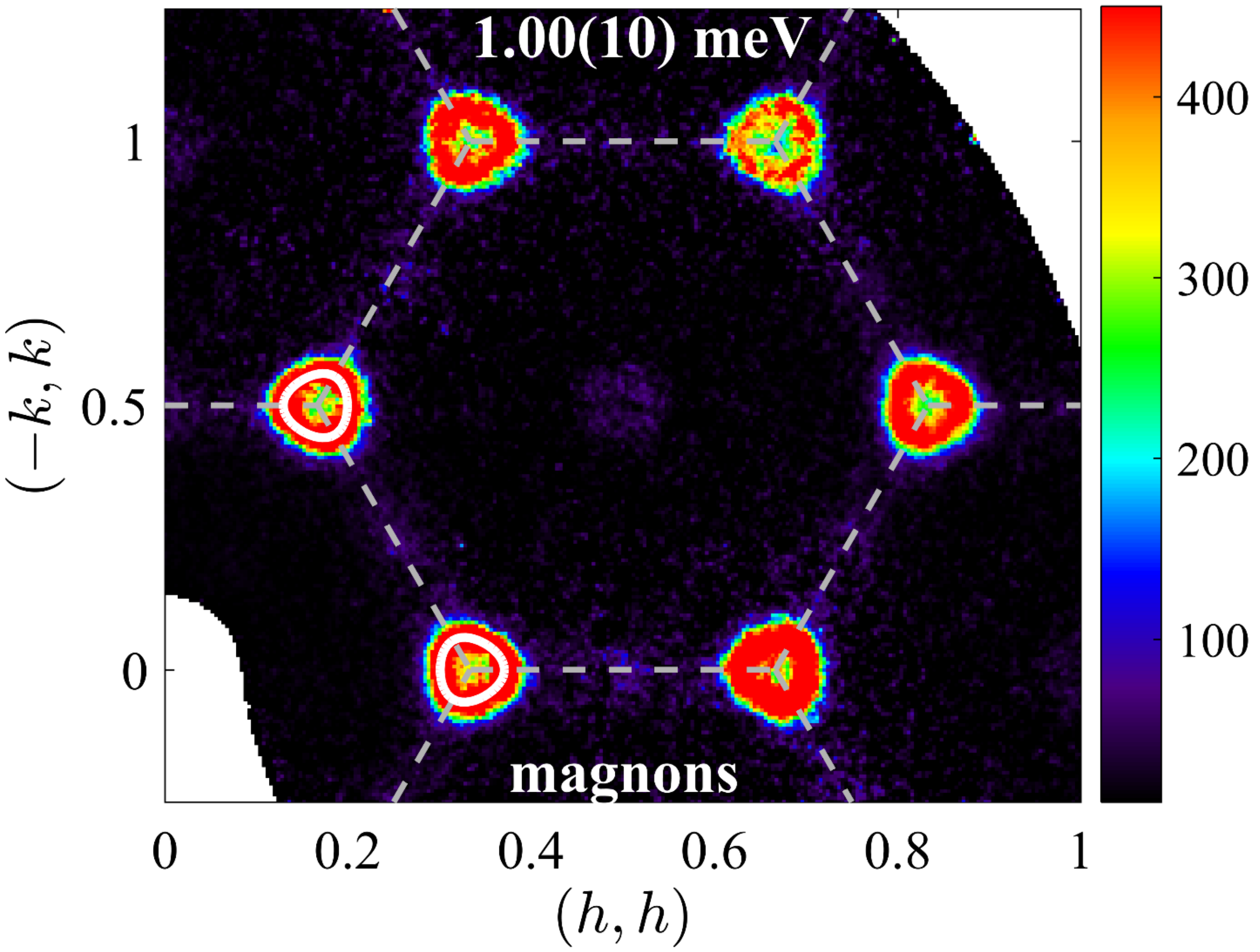
- continuum extends well beyond the top two-magnon cut-off
- starts close to spinwave cone (not with a large separation) and has additional higher-energy structure



# Intensity modulations in the continuum scattering



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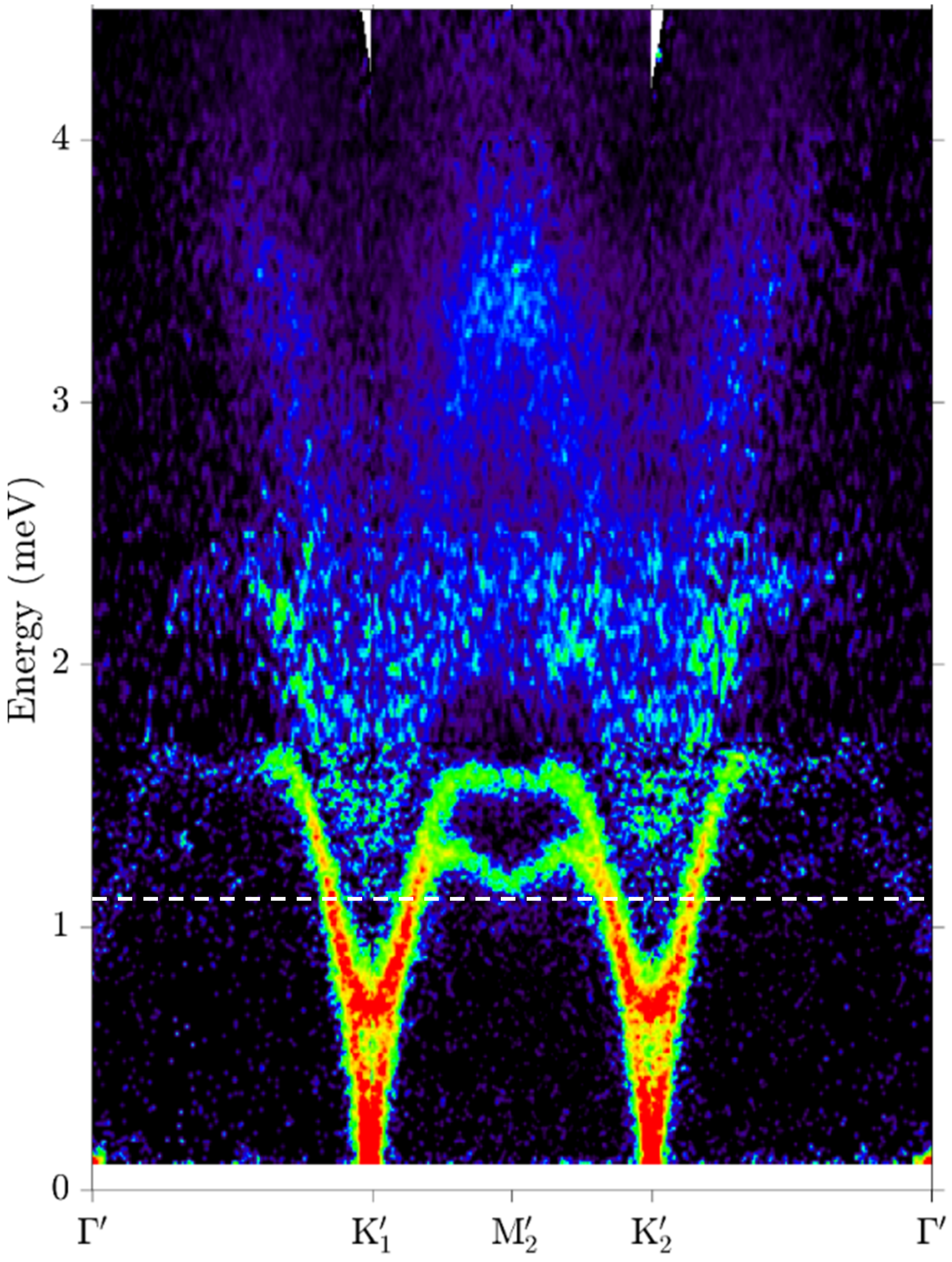
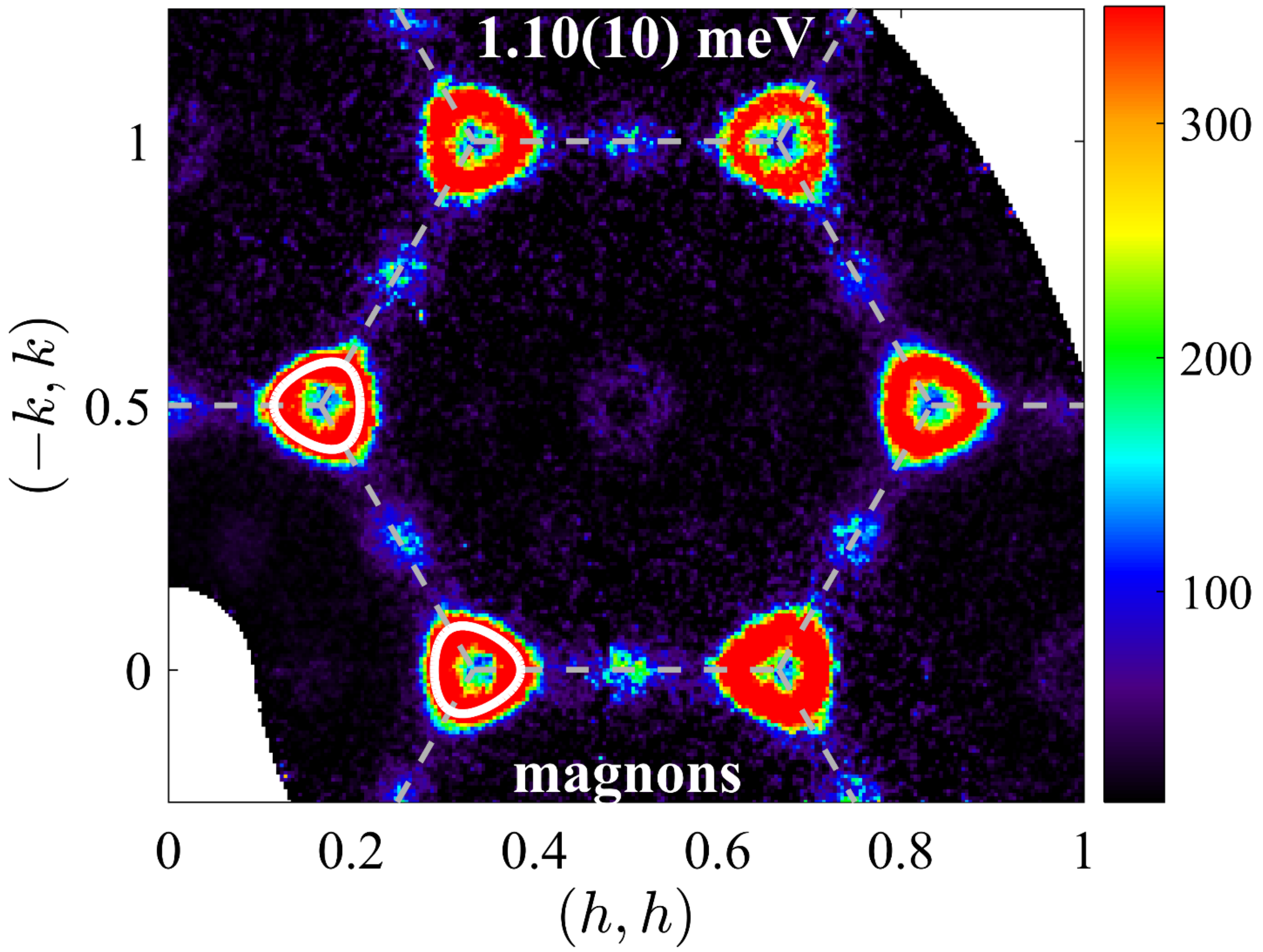
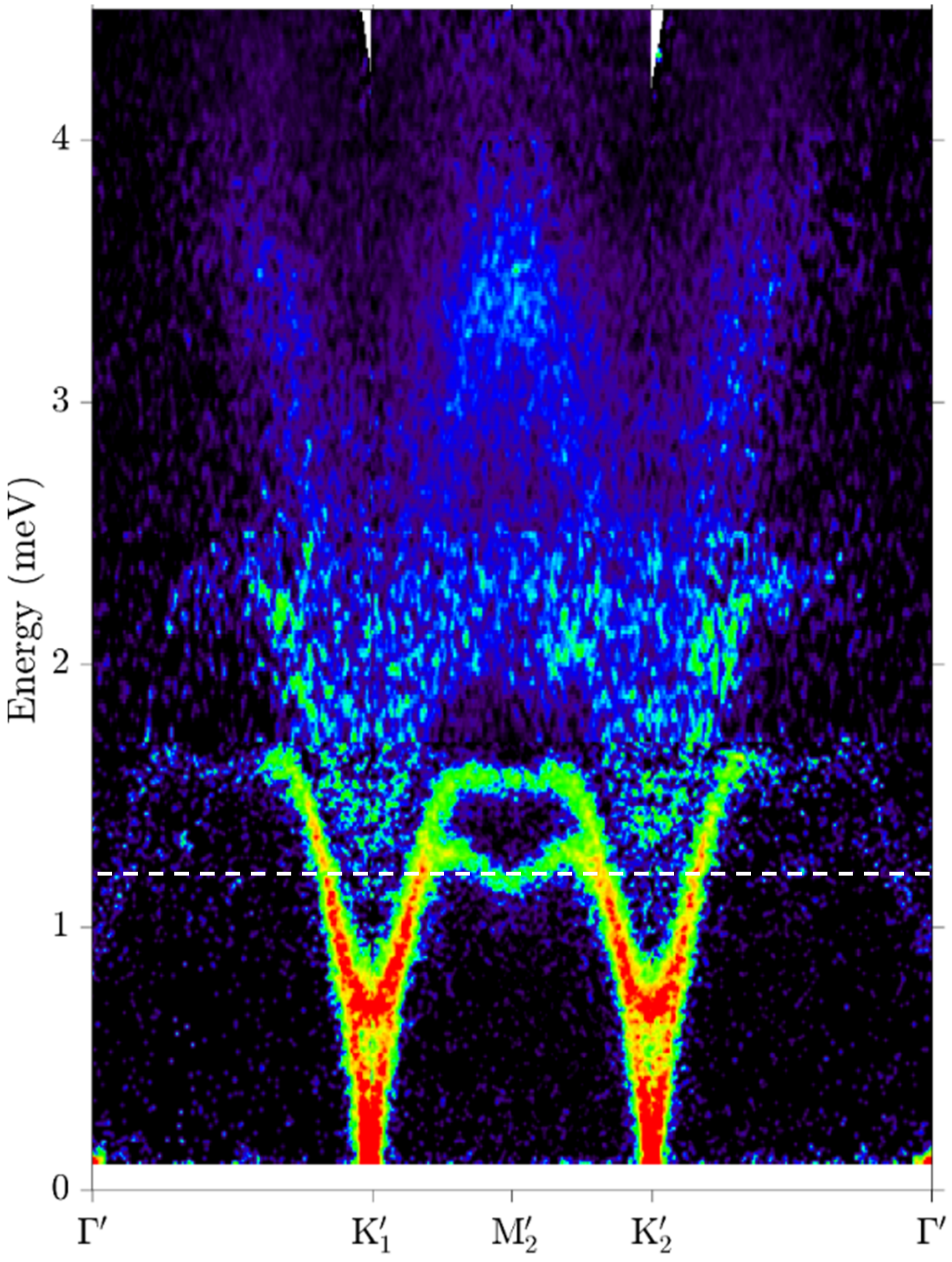
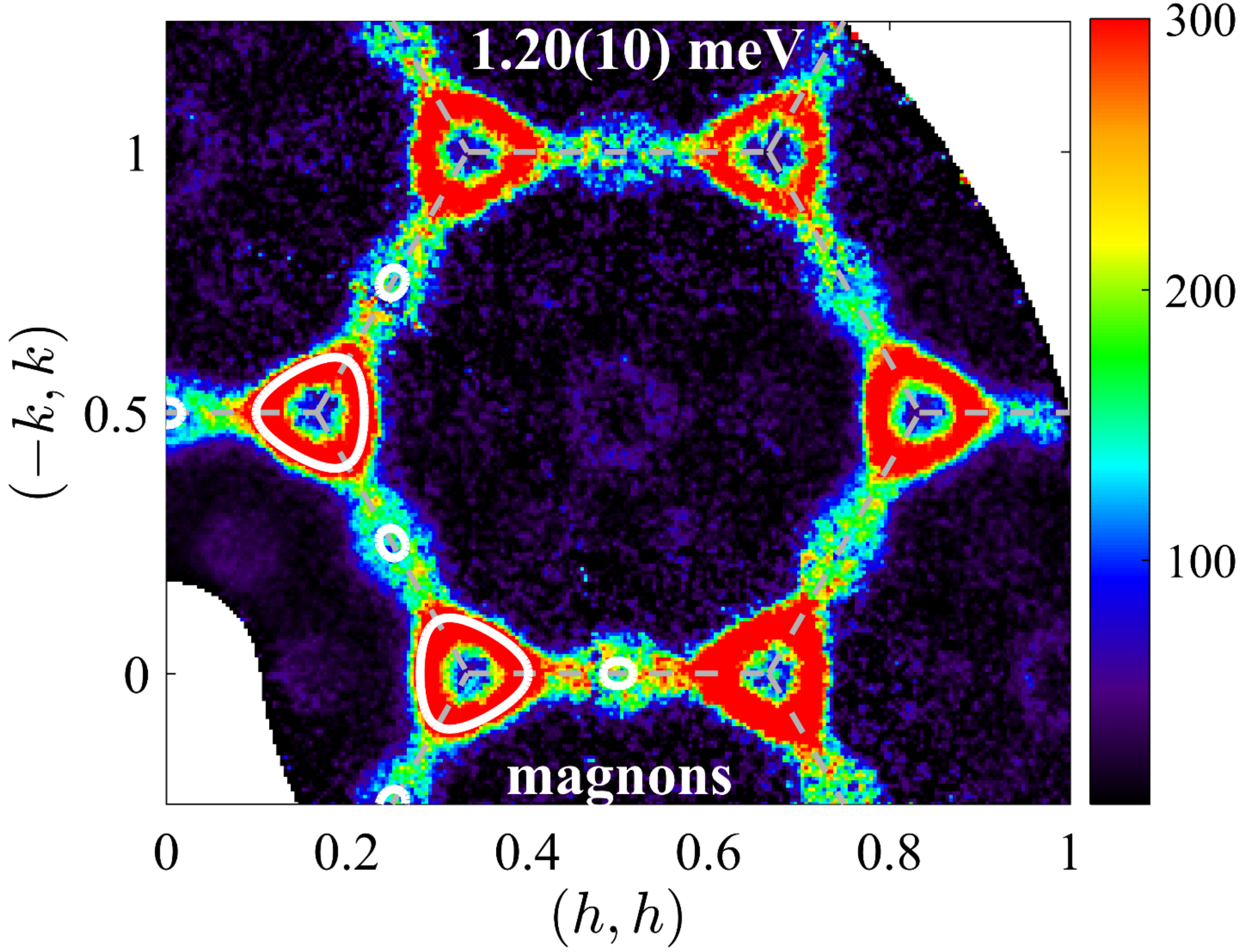


# Intensity modulations in the continuum scattering



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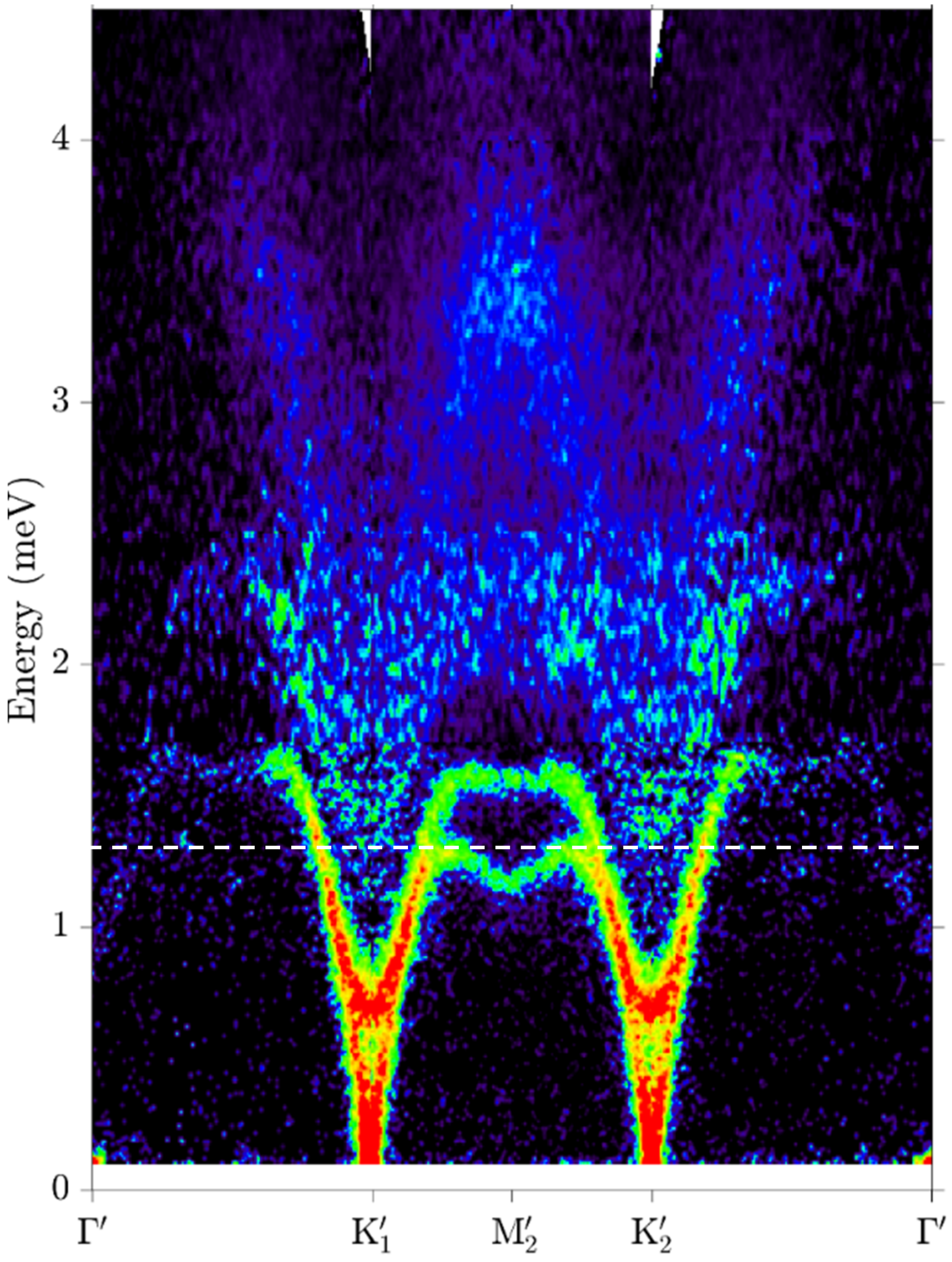


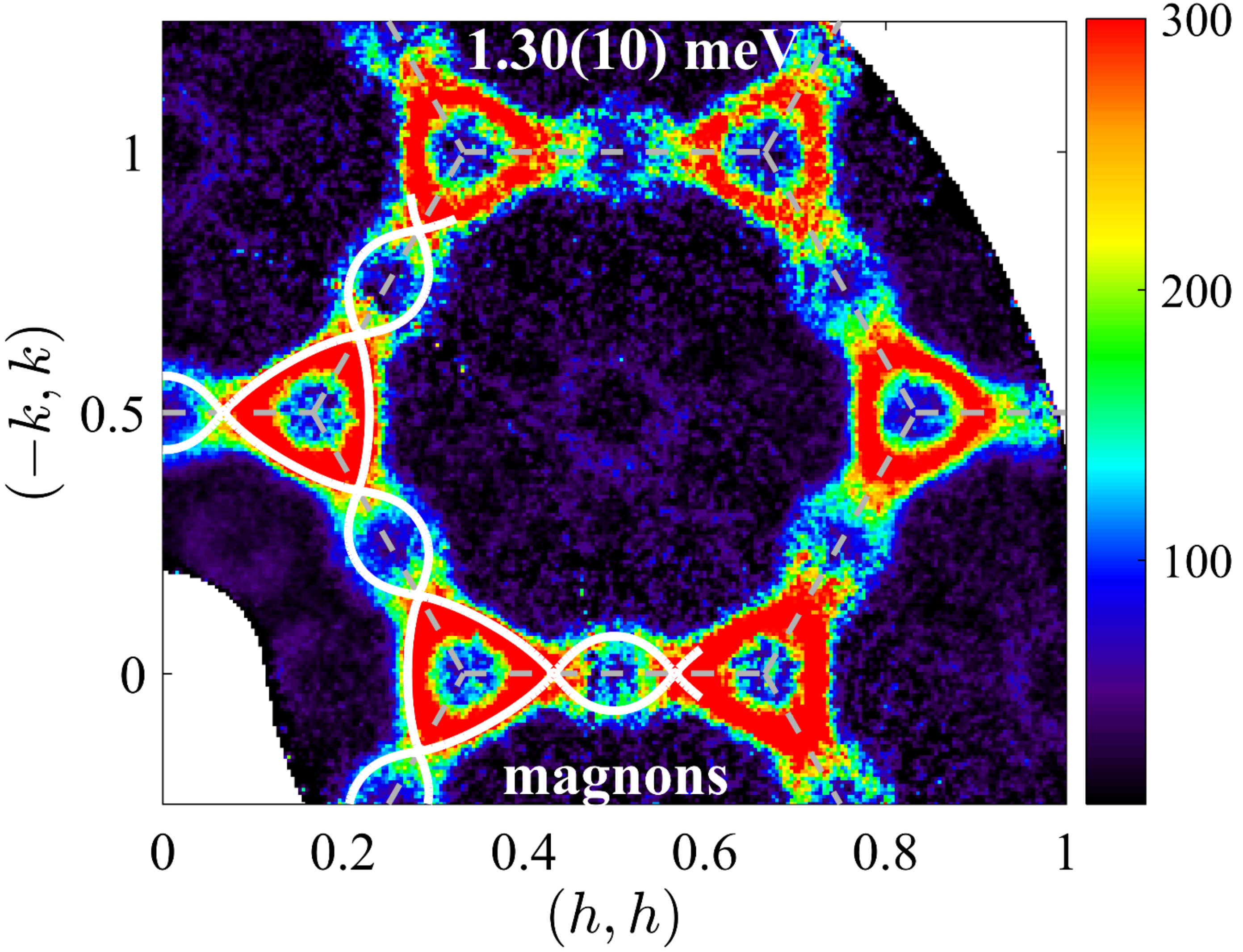

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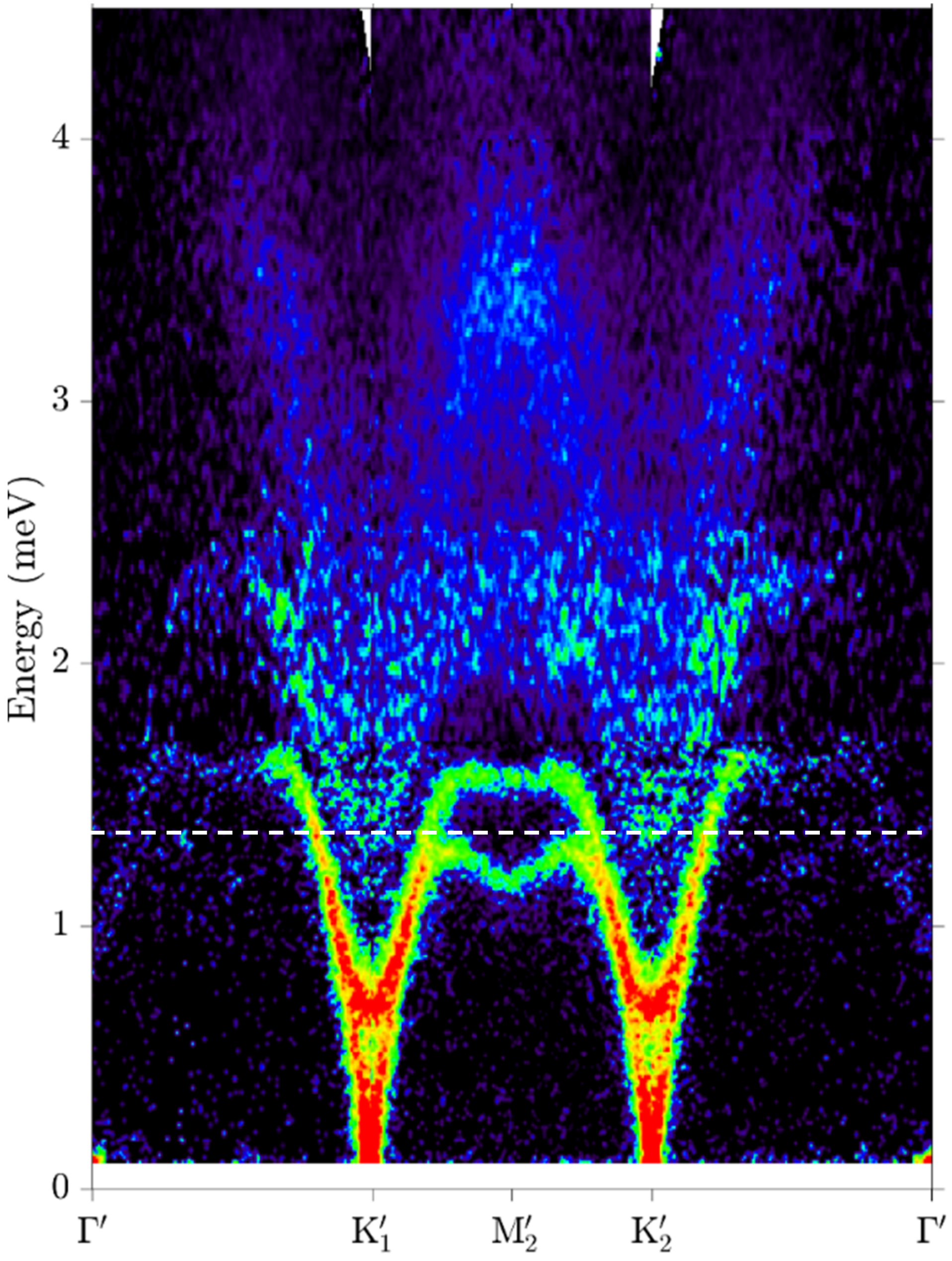


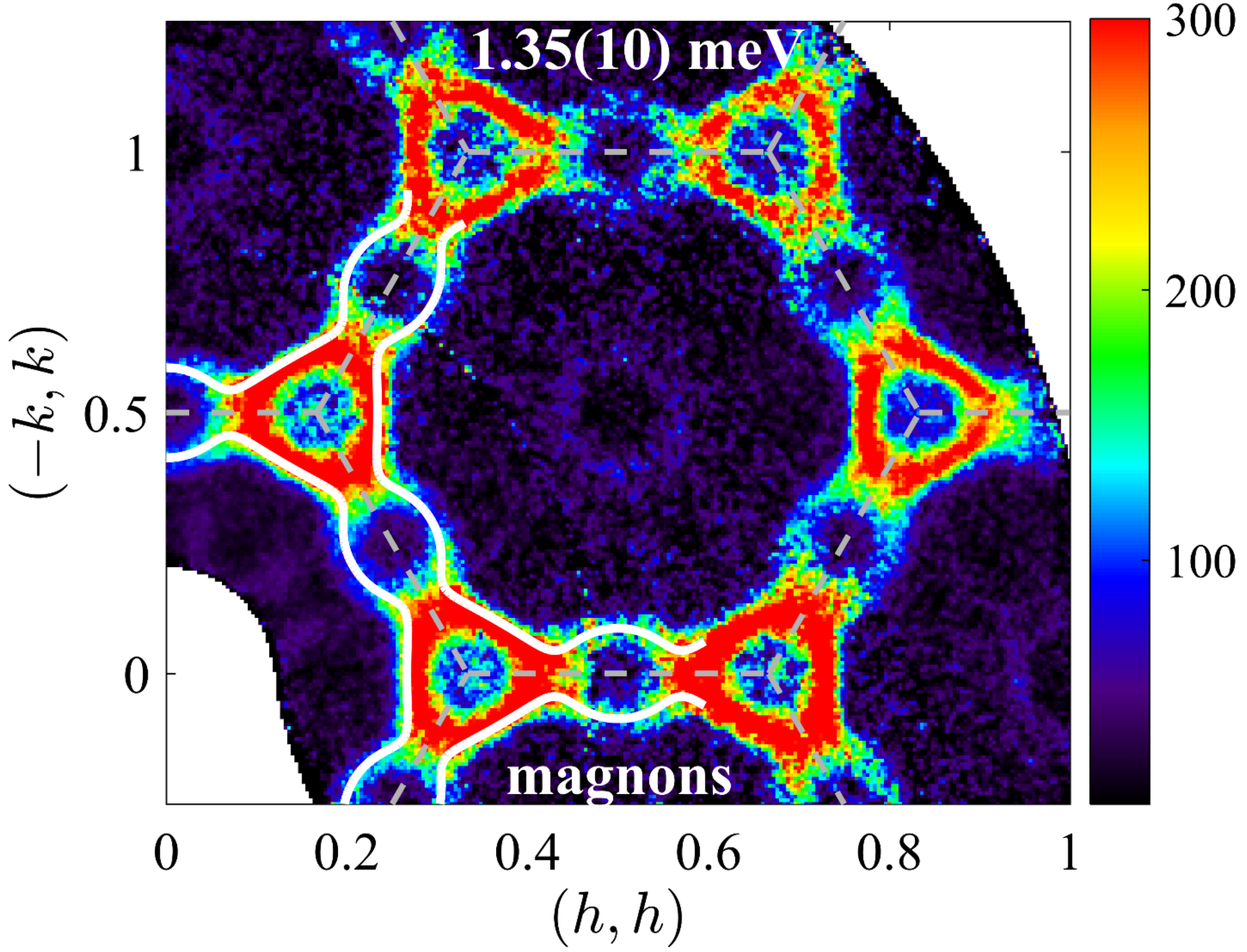

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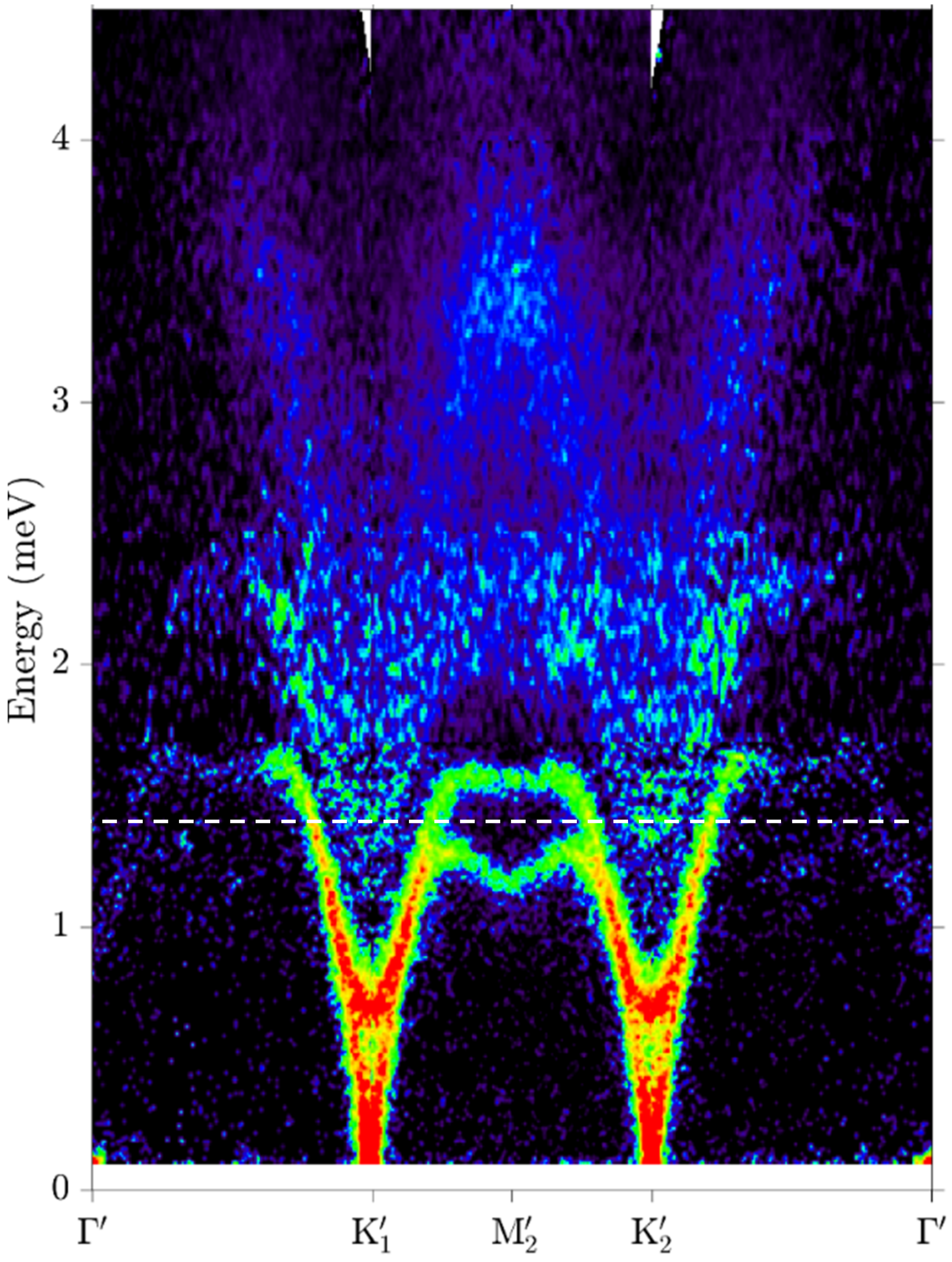


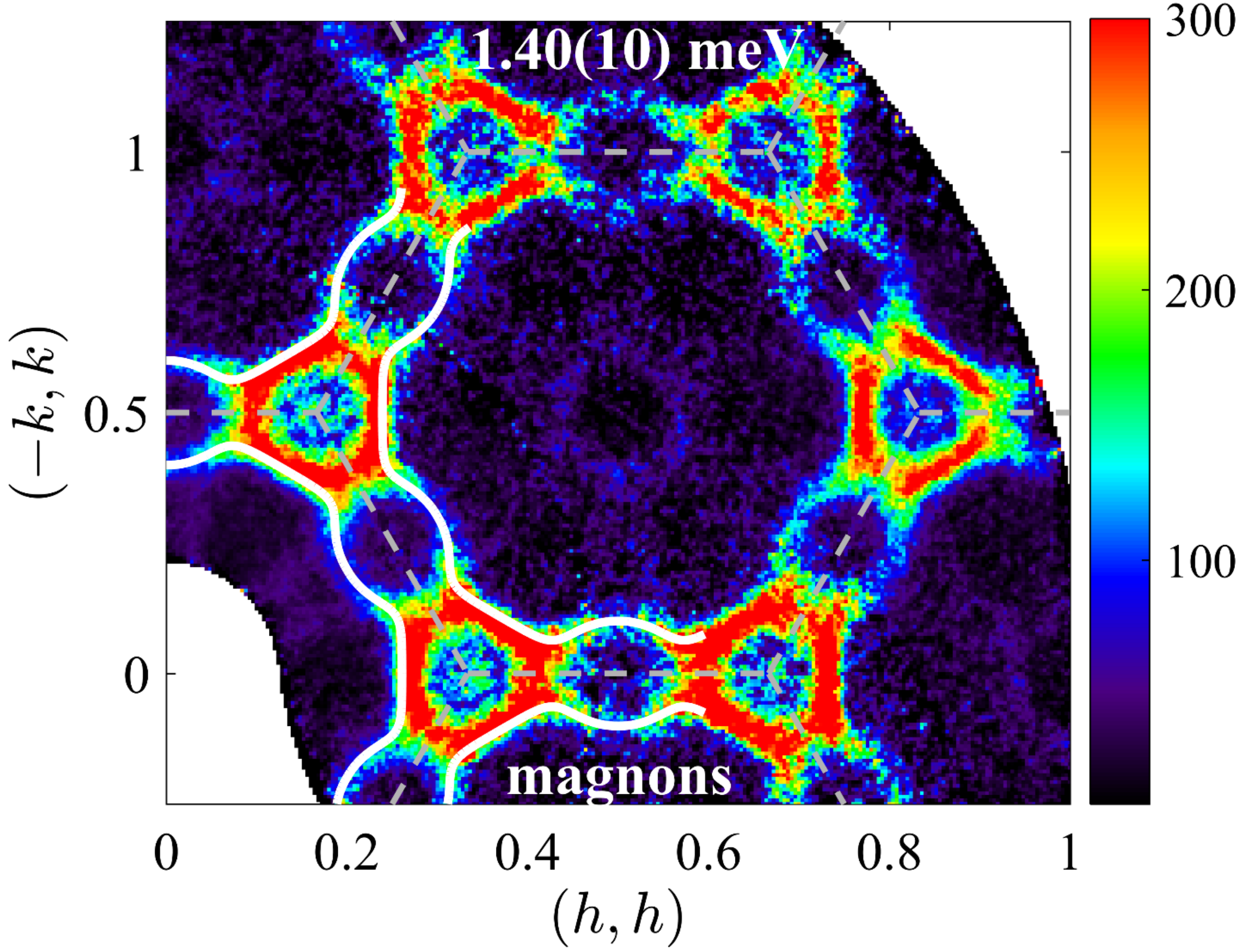

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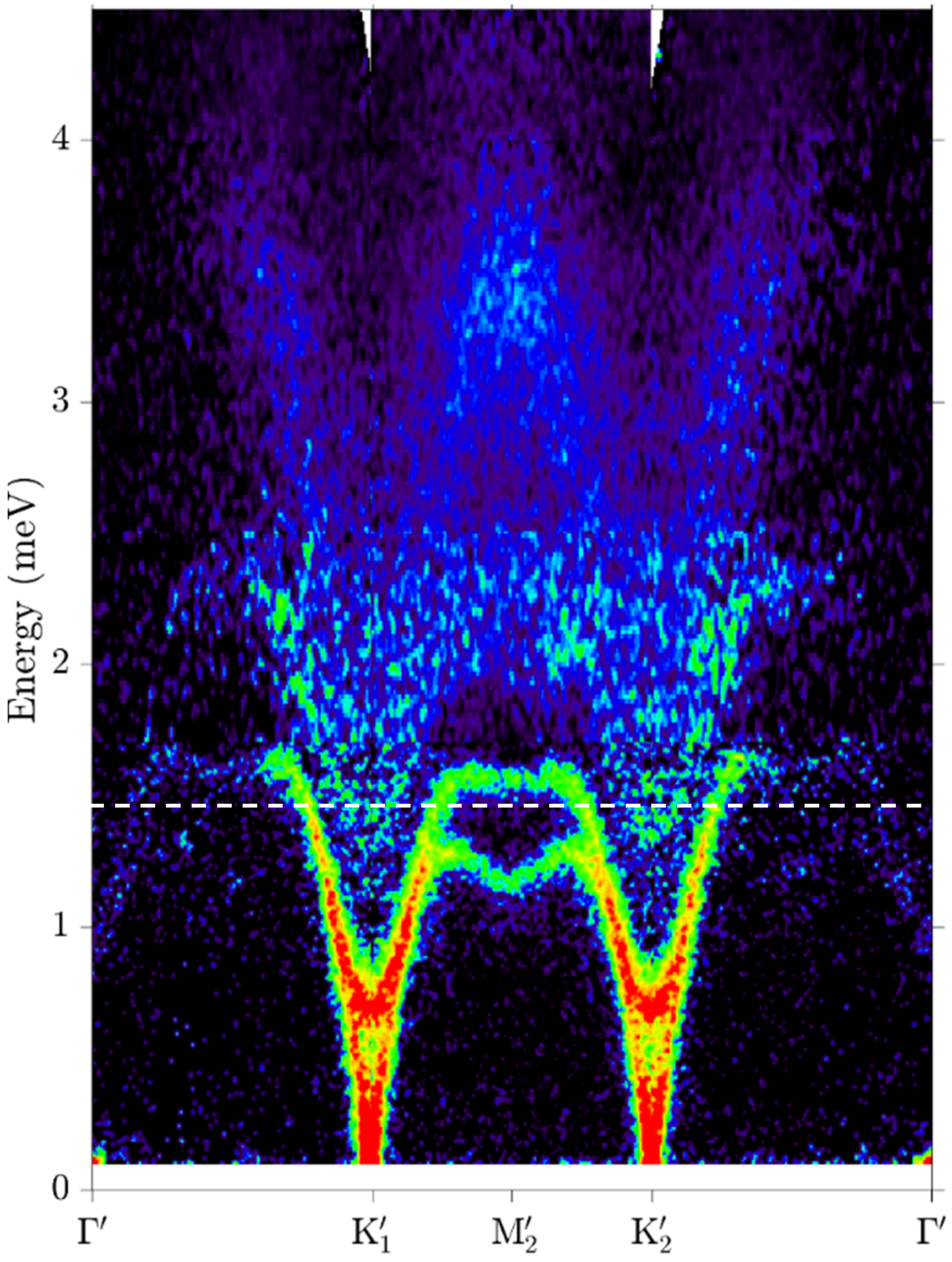


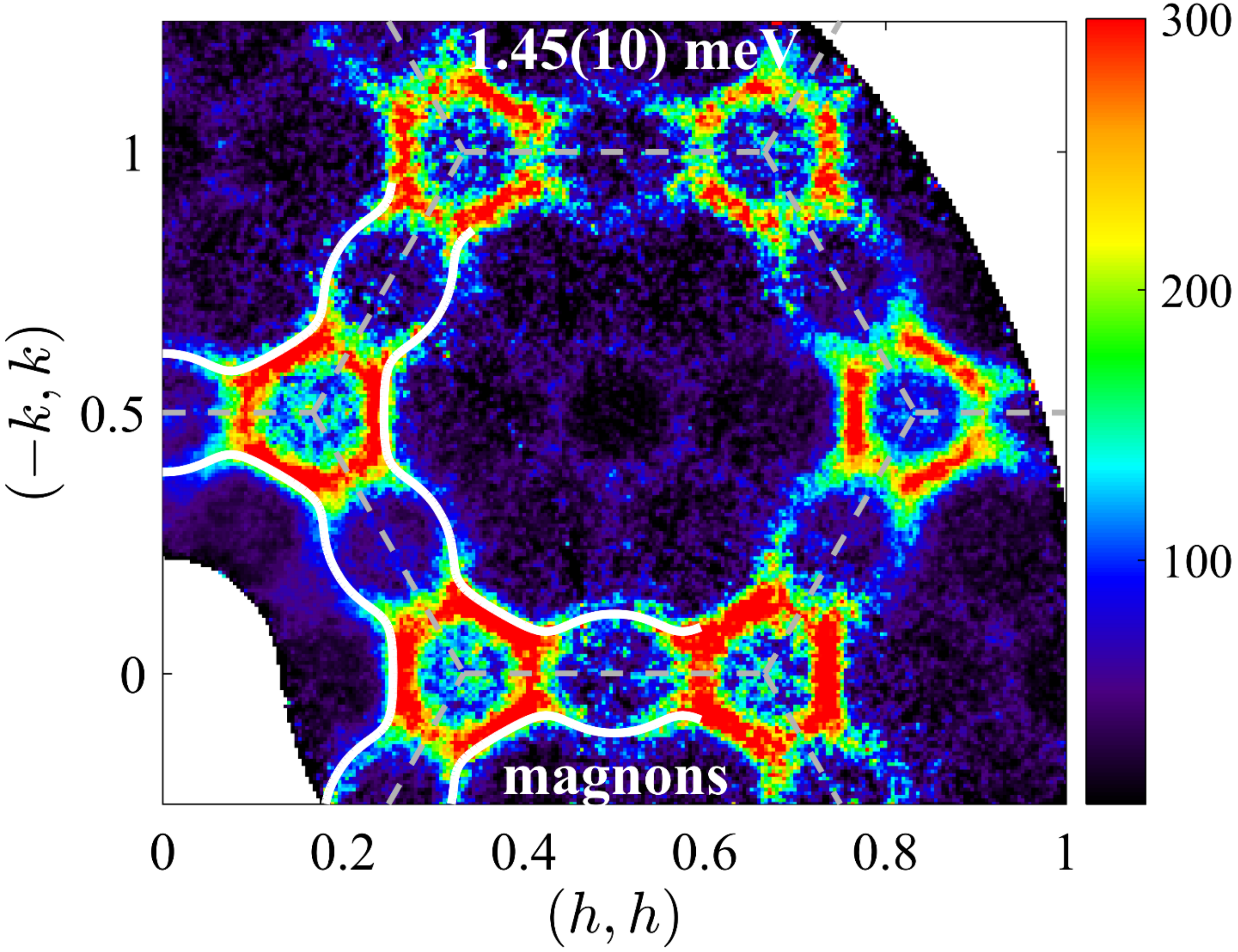

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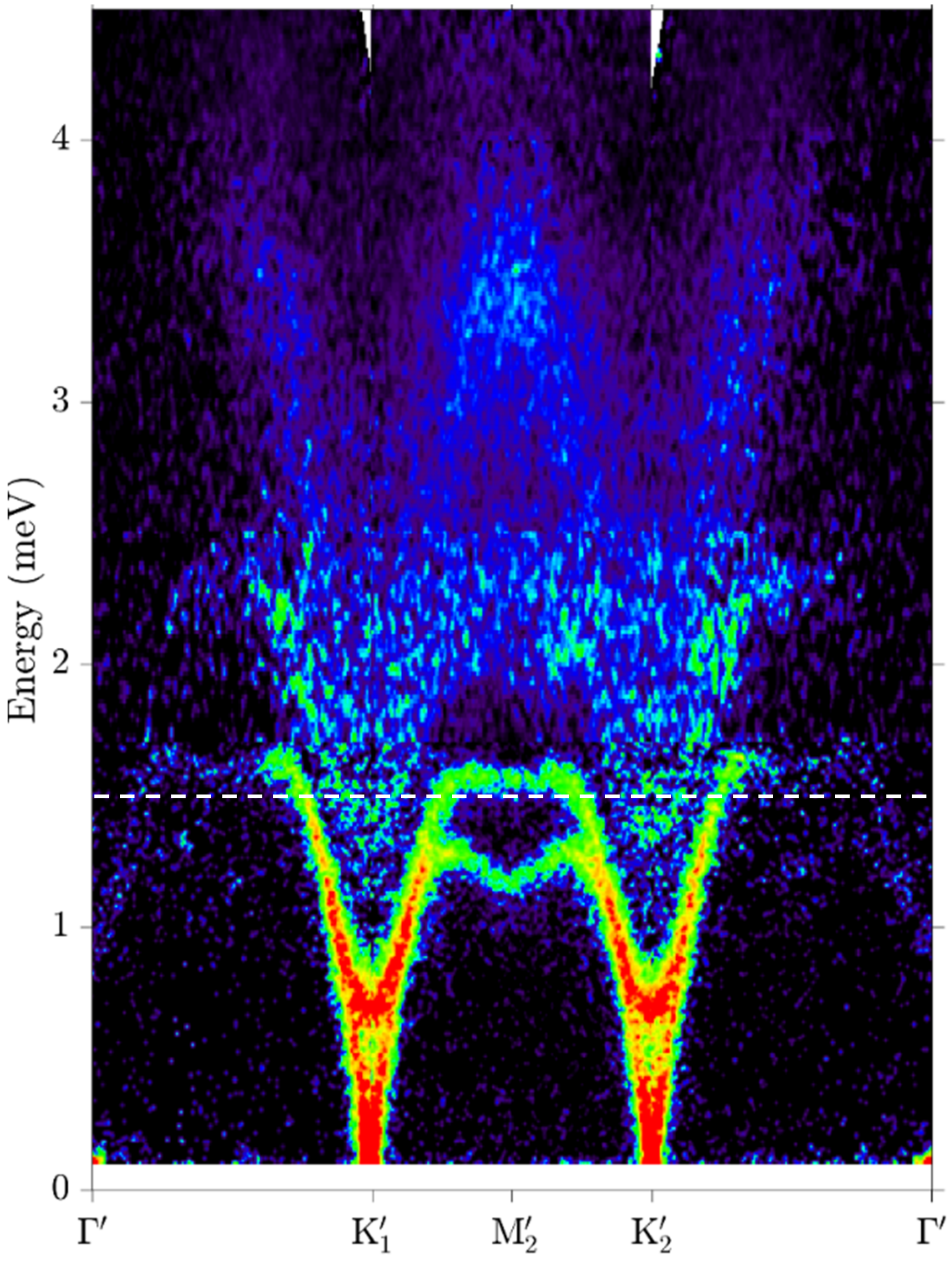


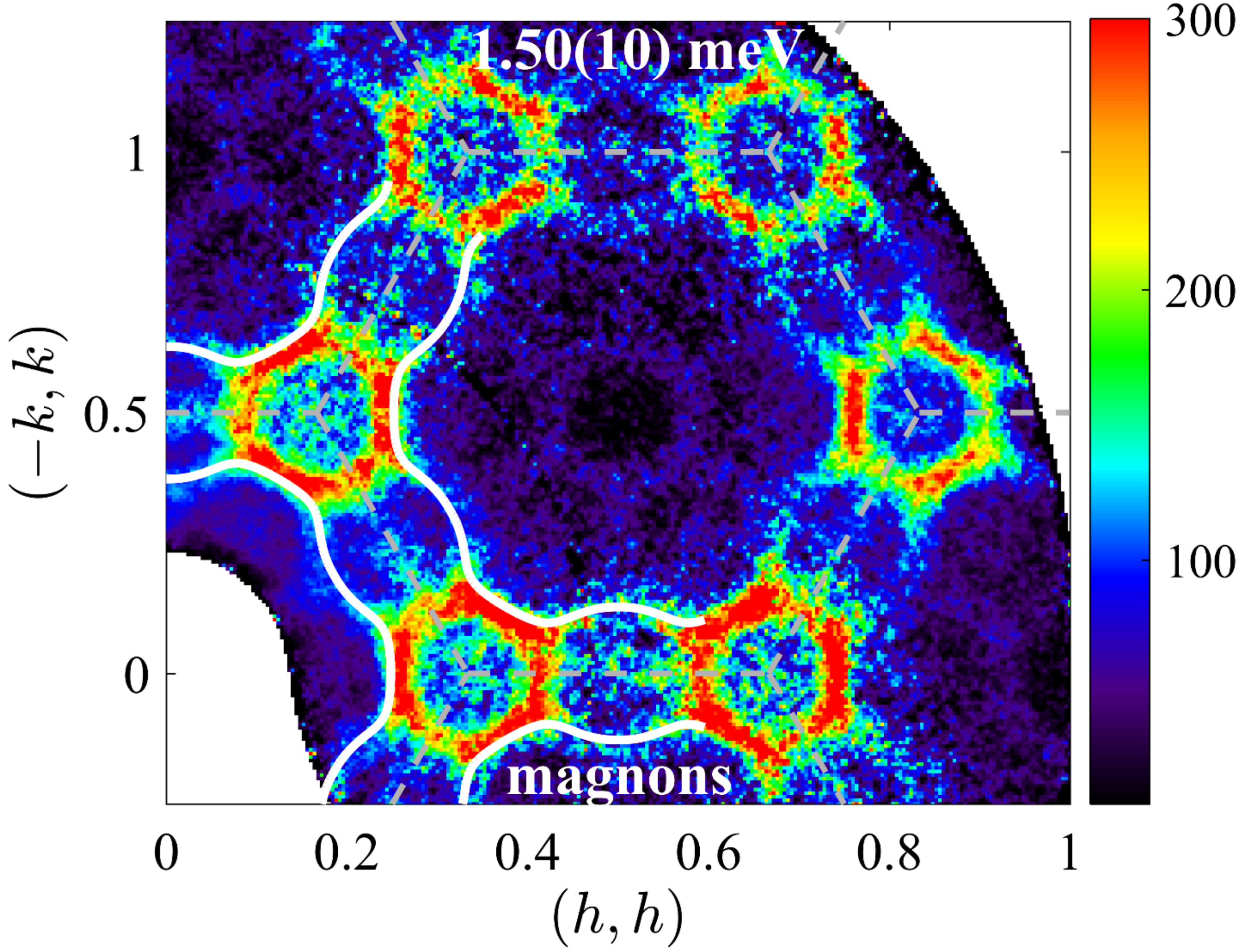

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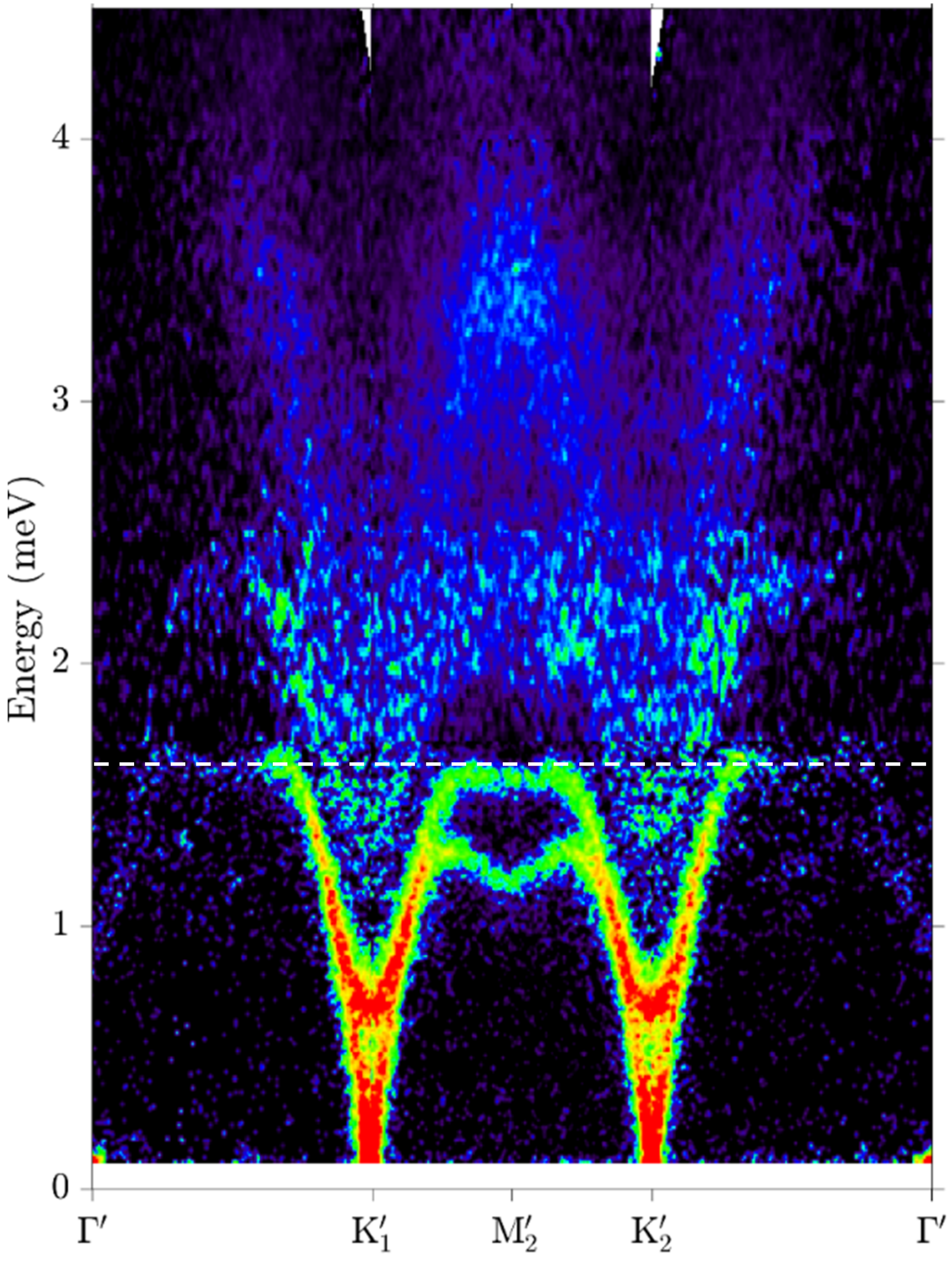
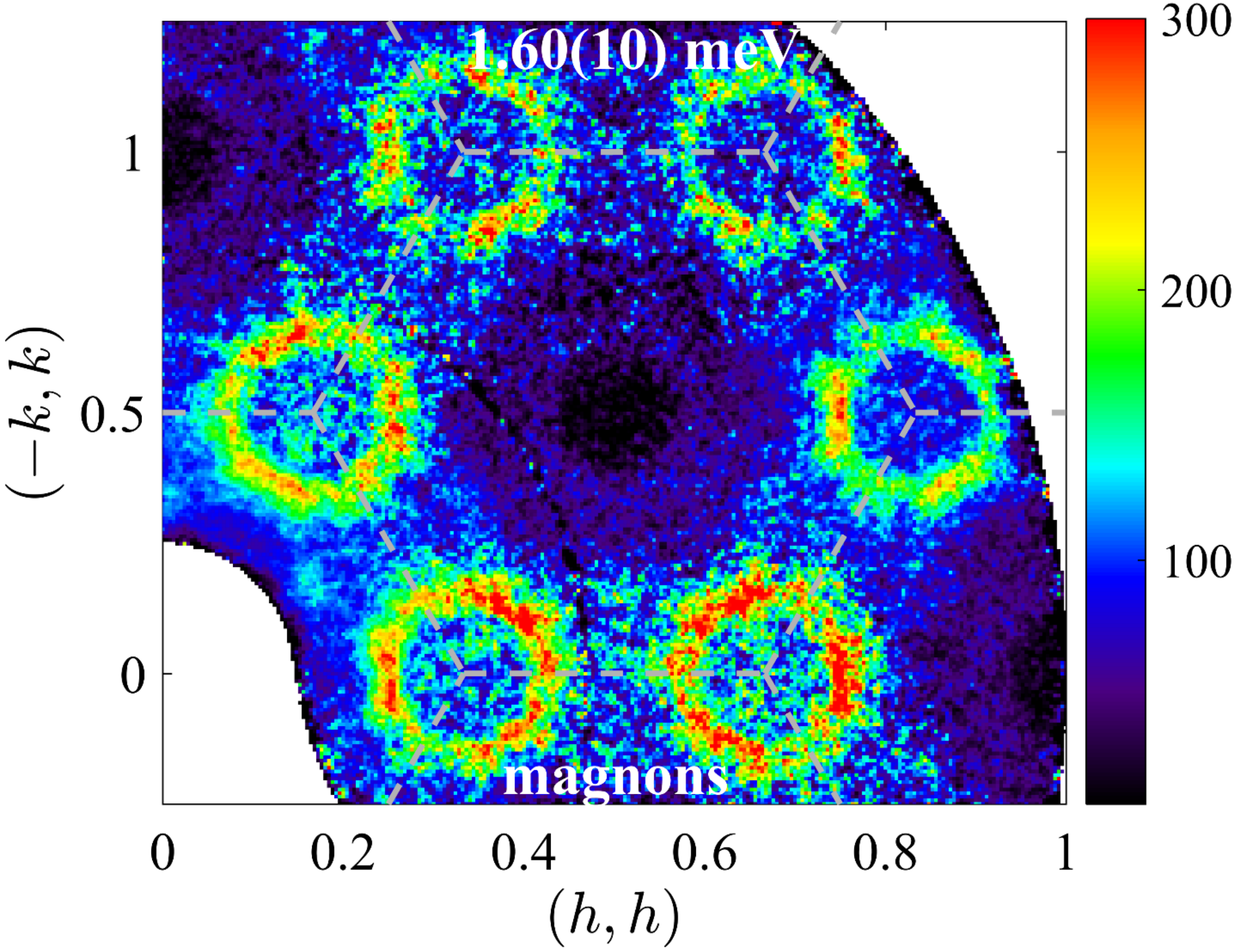
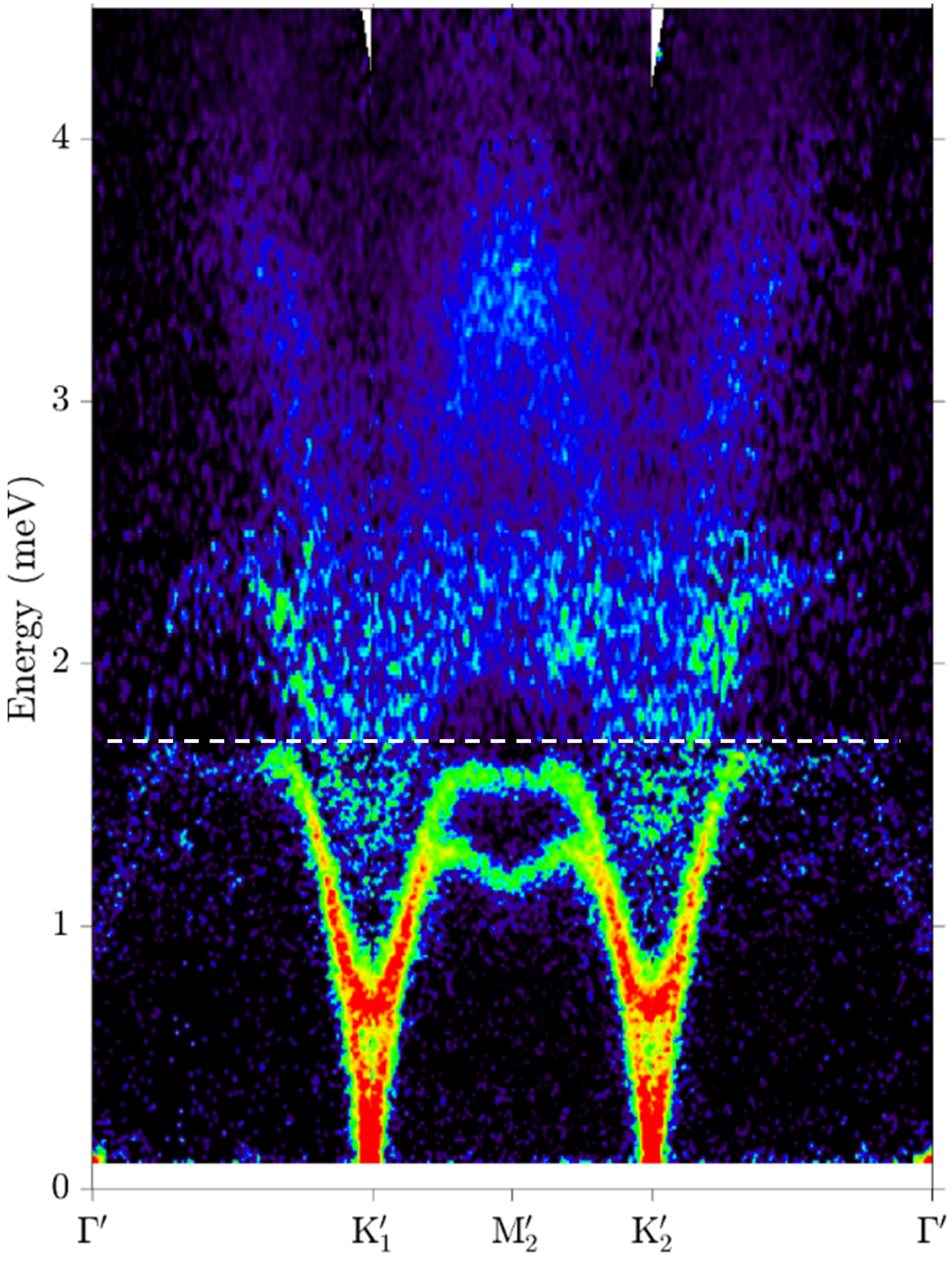
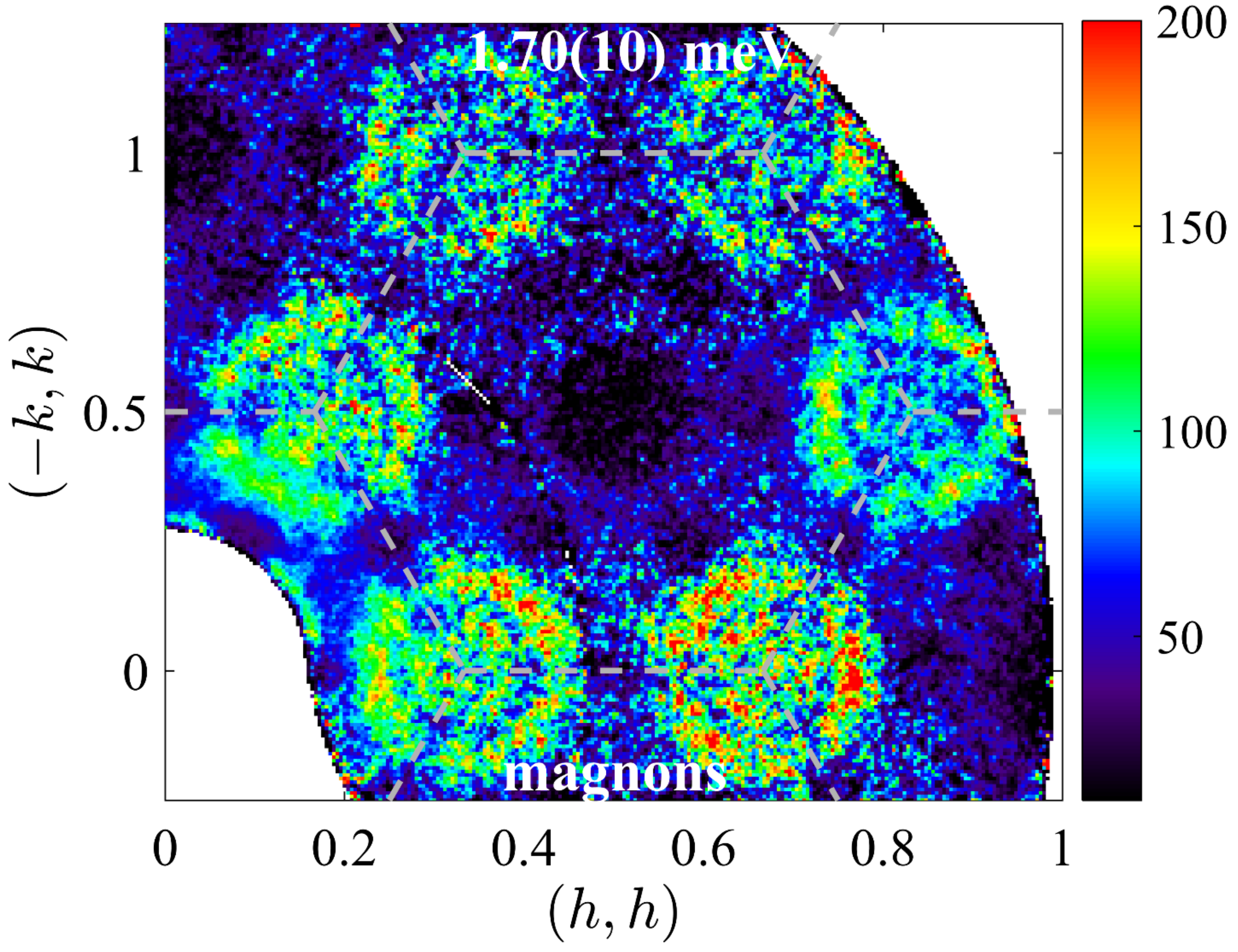


# Intensity modulations in the continuum scattering

- **continuum intensity is highly structured** with patterns of rings, triangles and hexagons appearing at various energies



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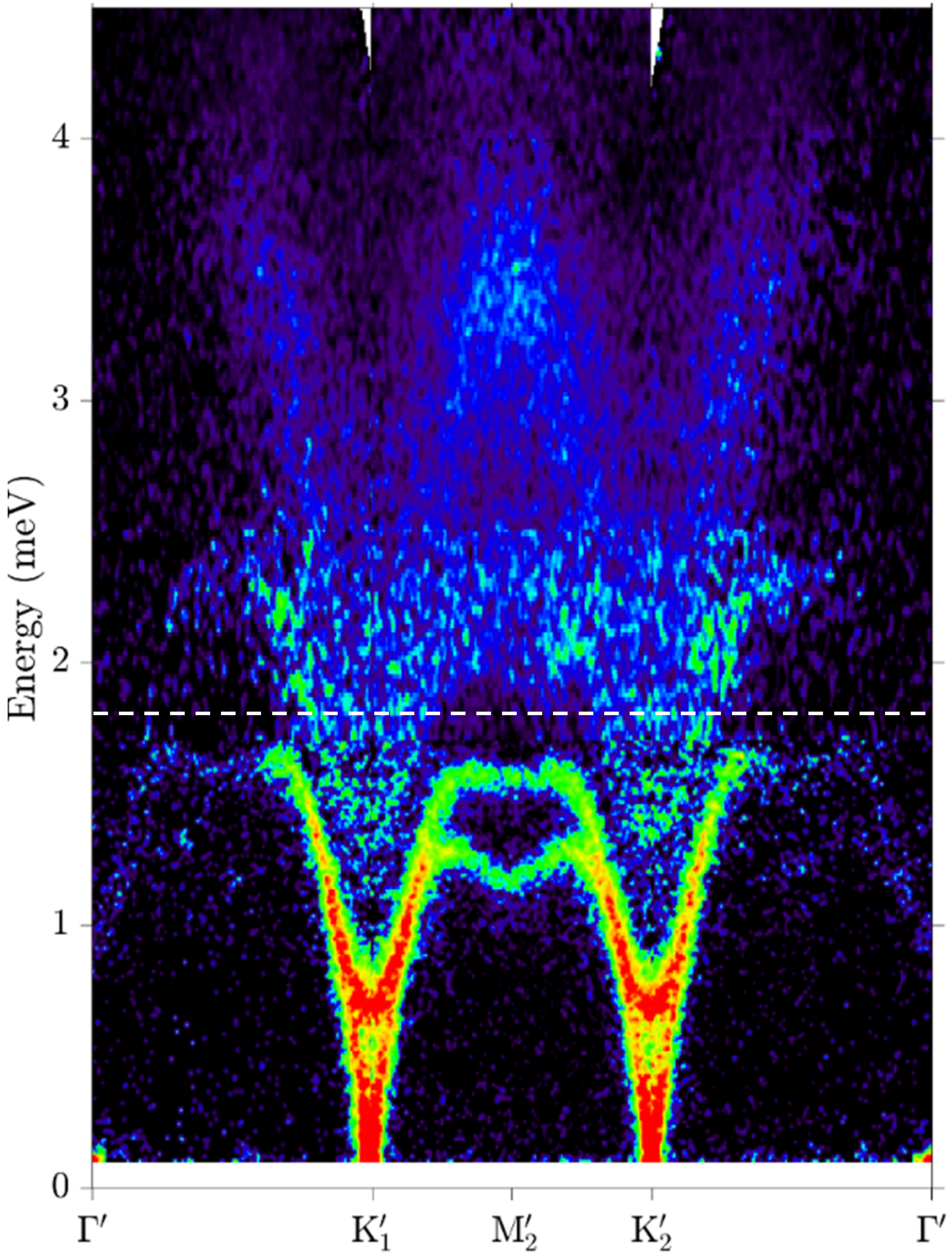


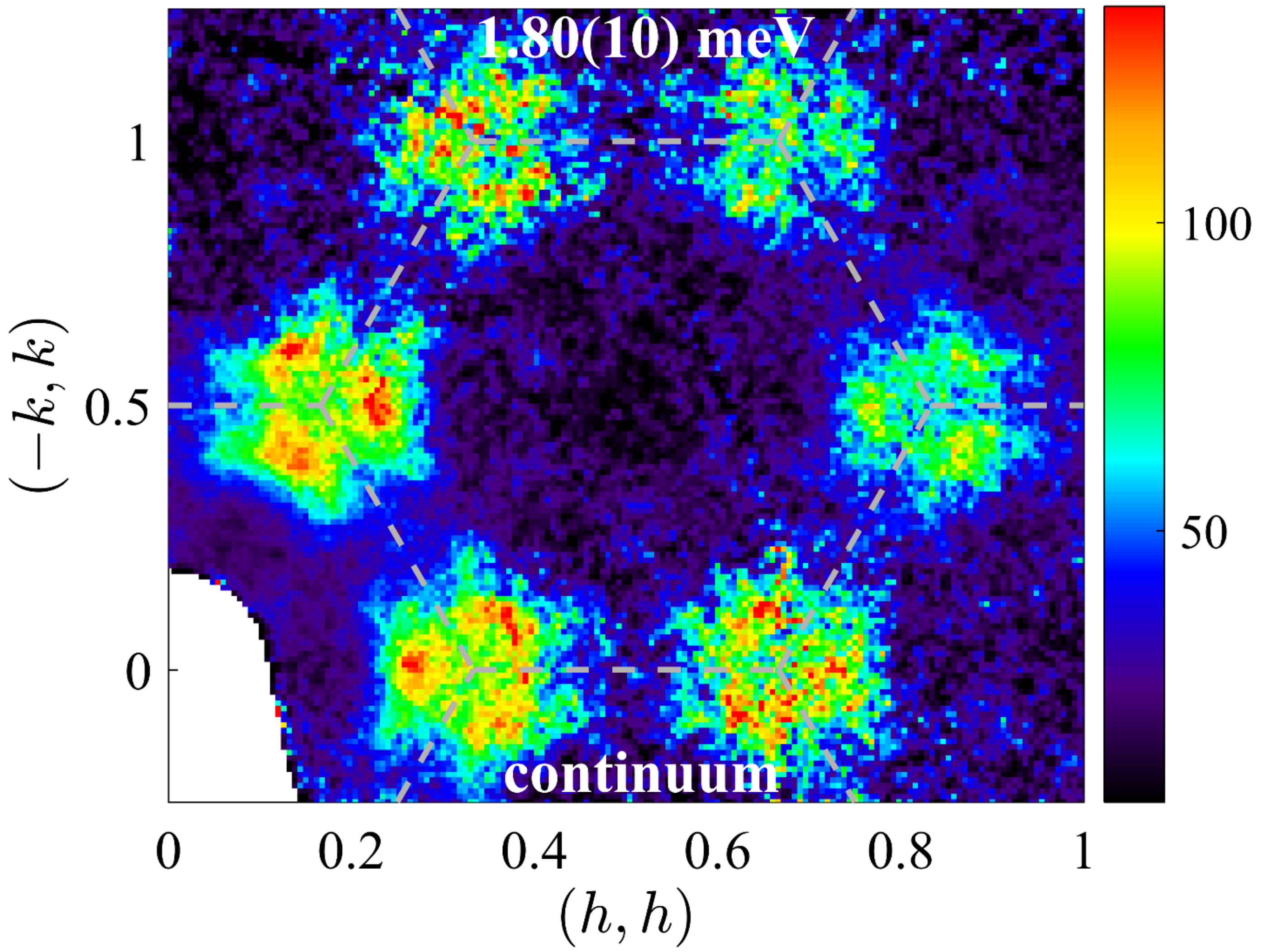

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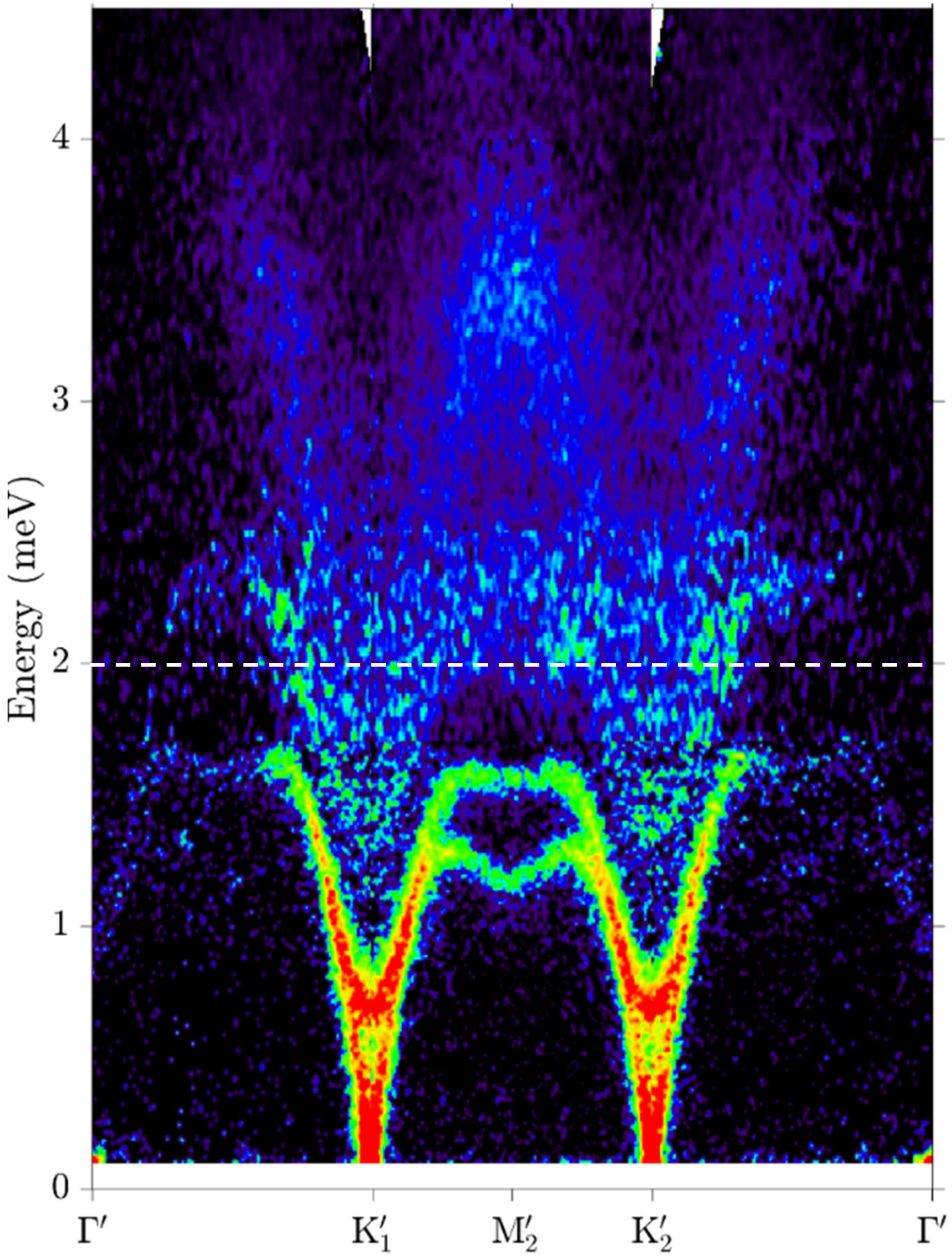


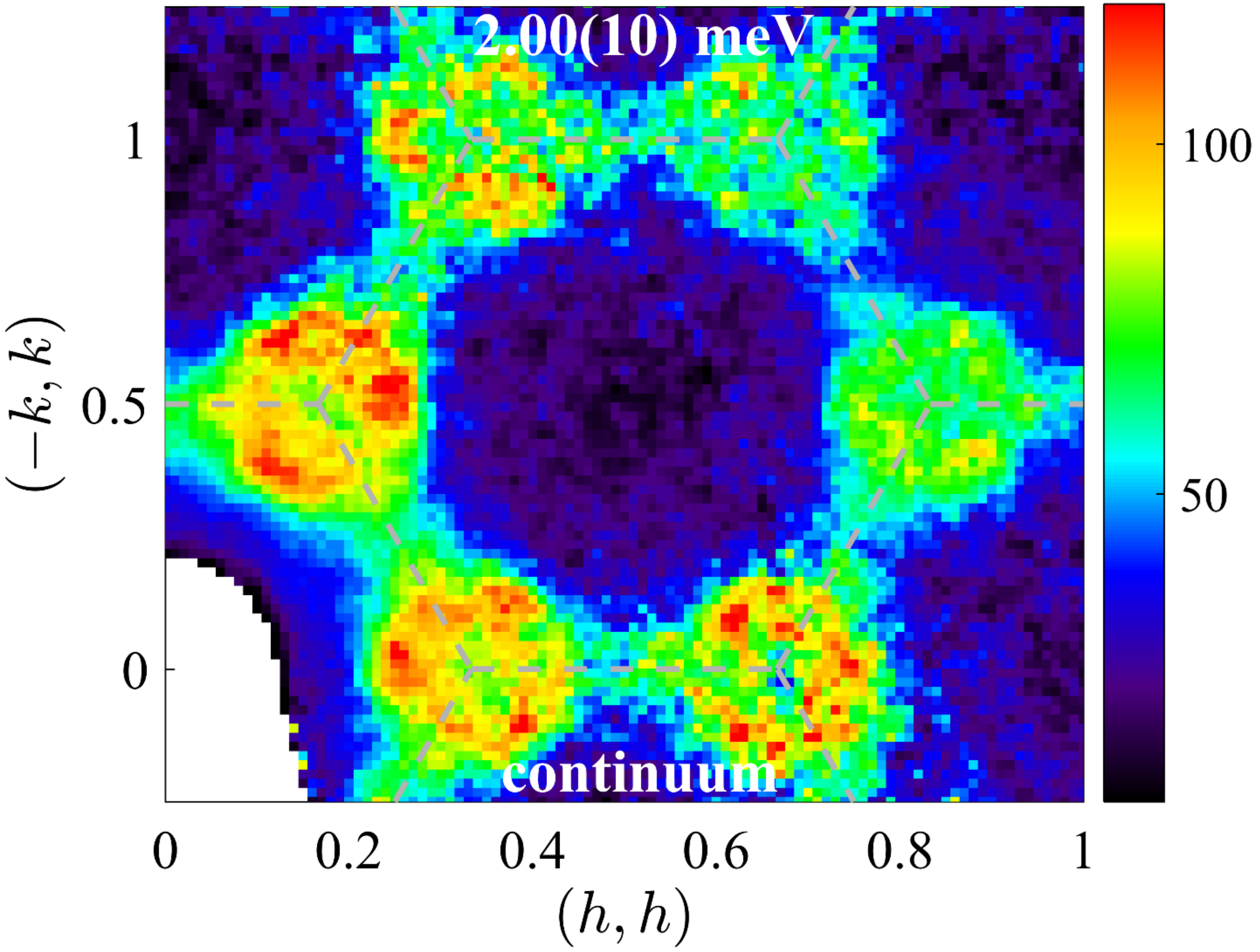

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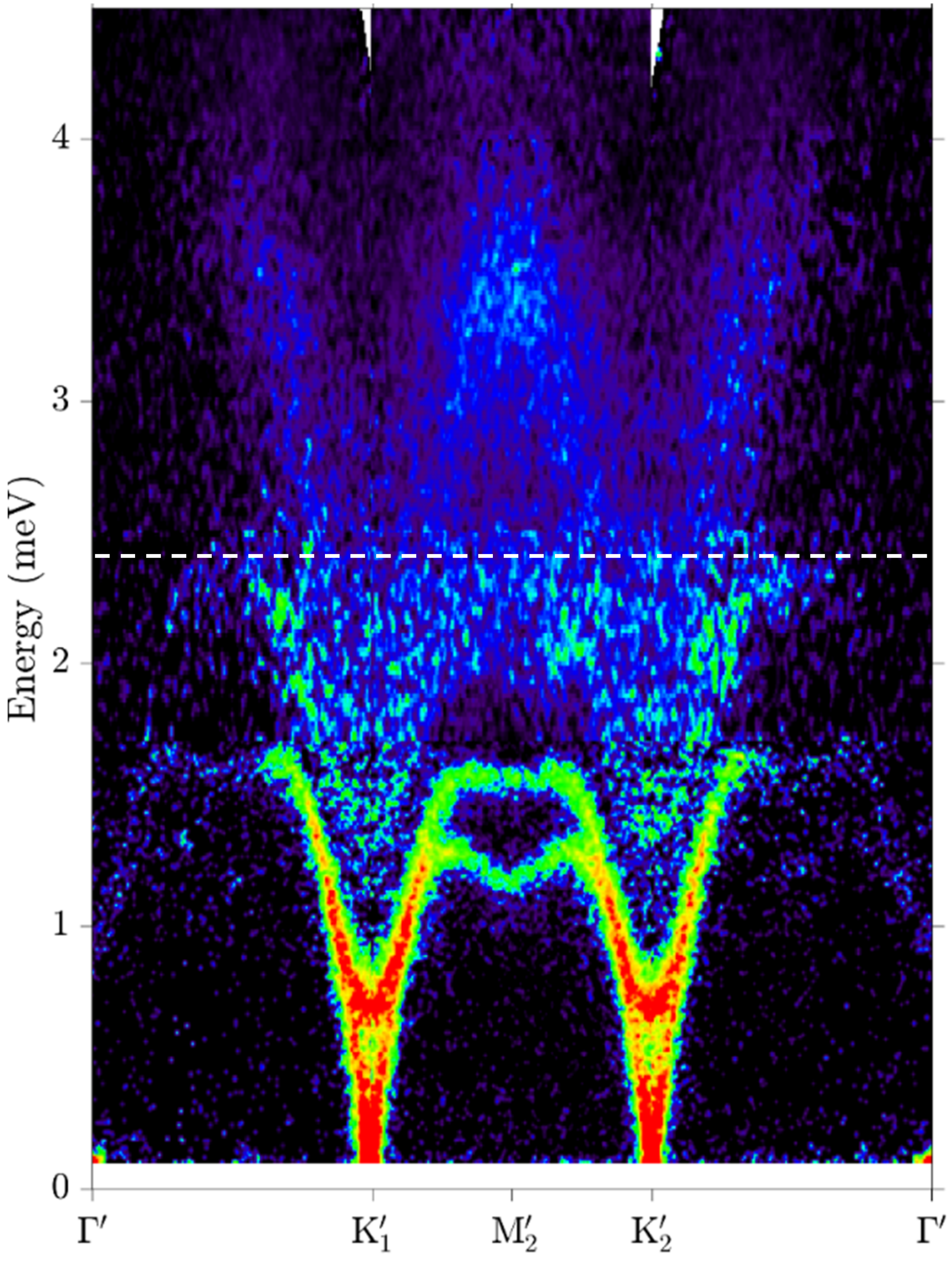


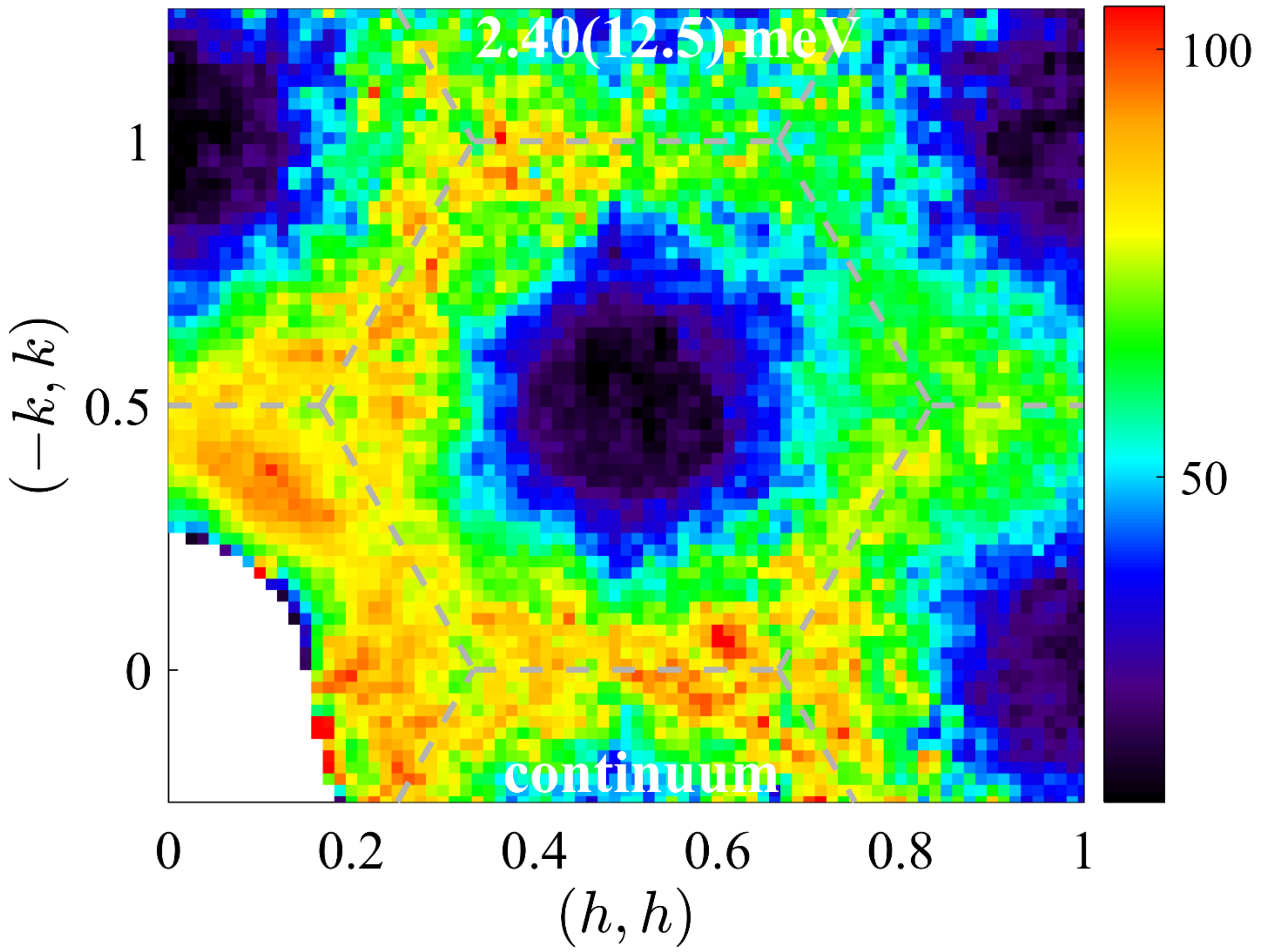

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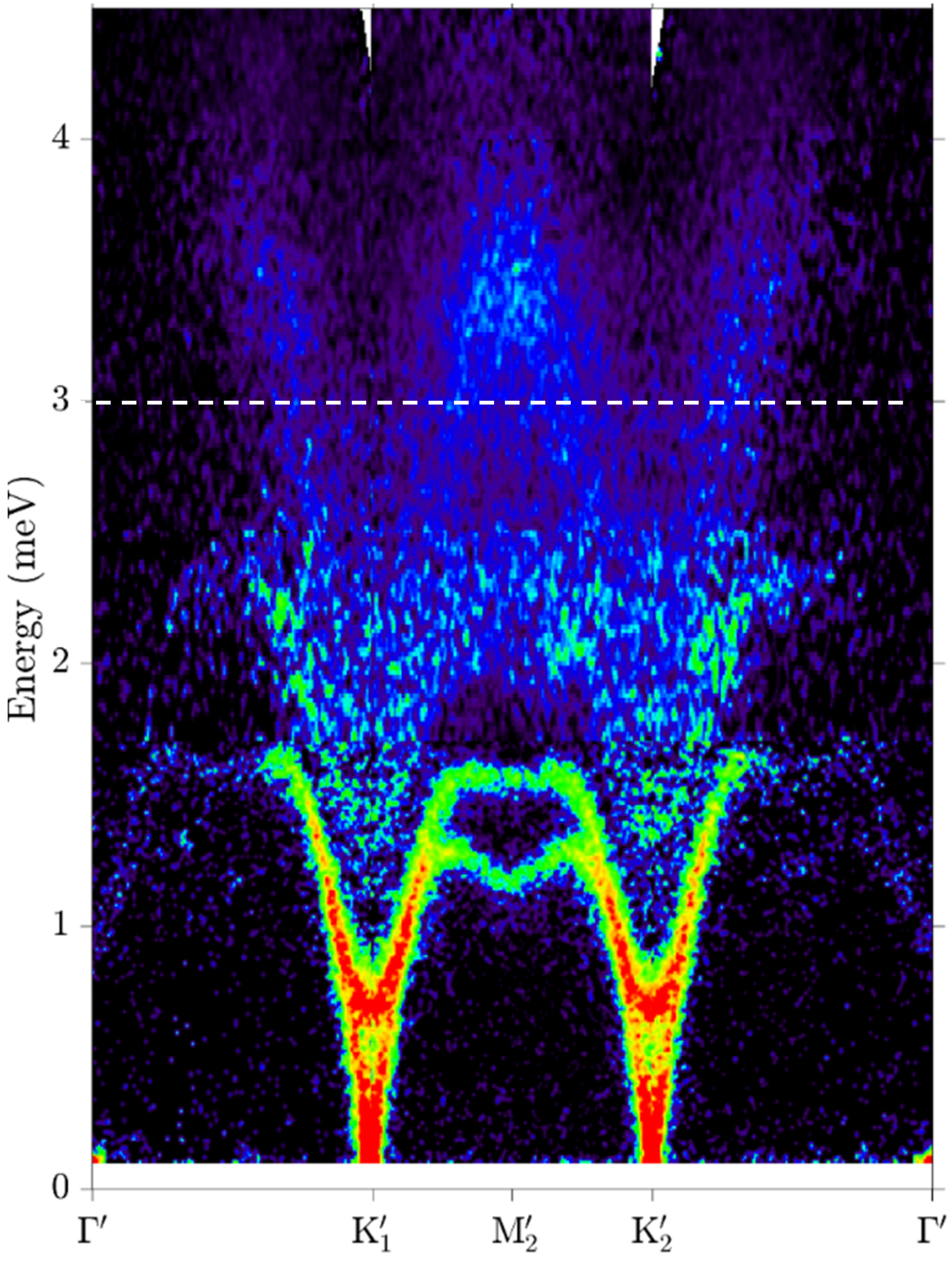


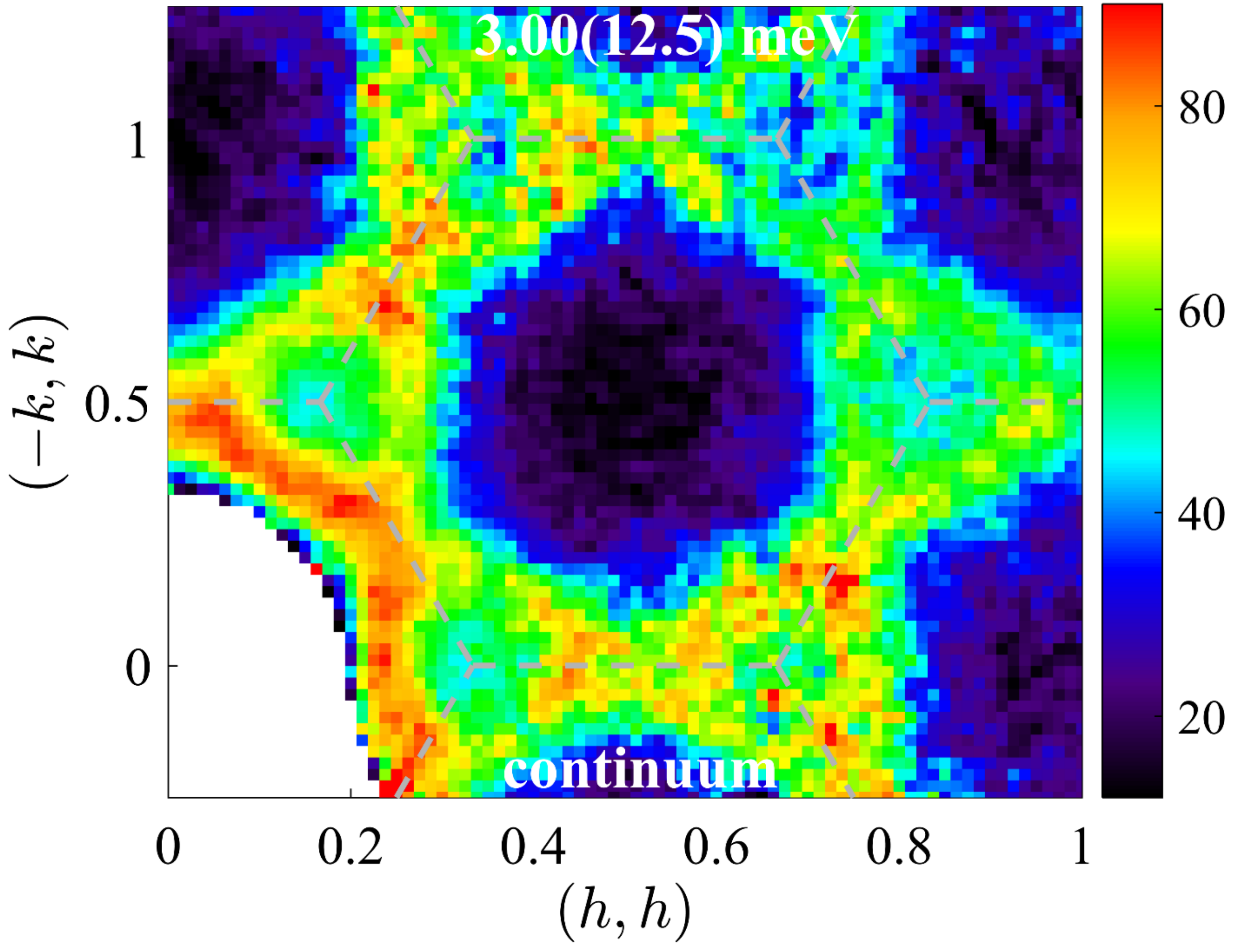

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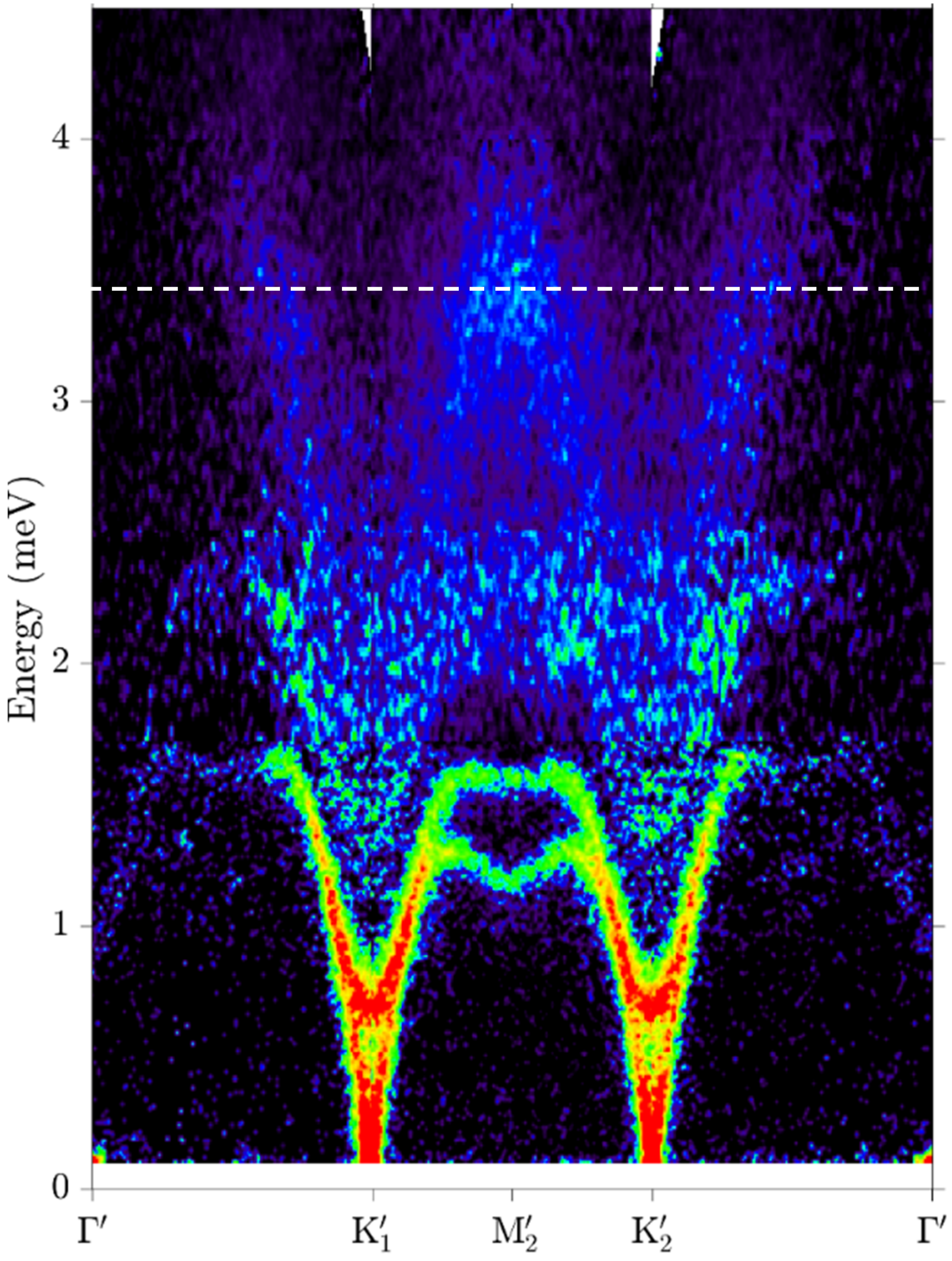
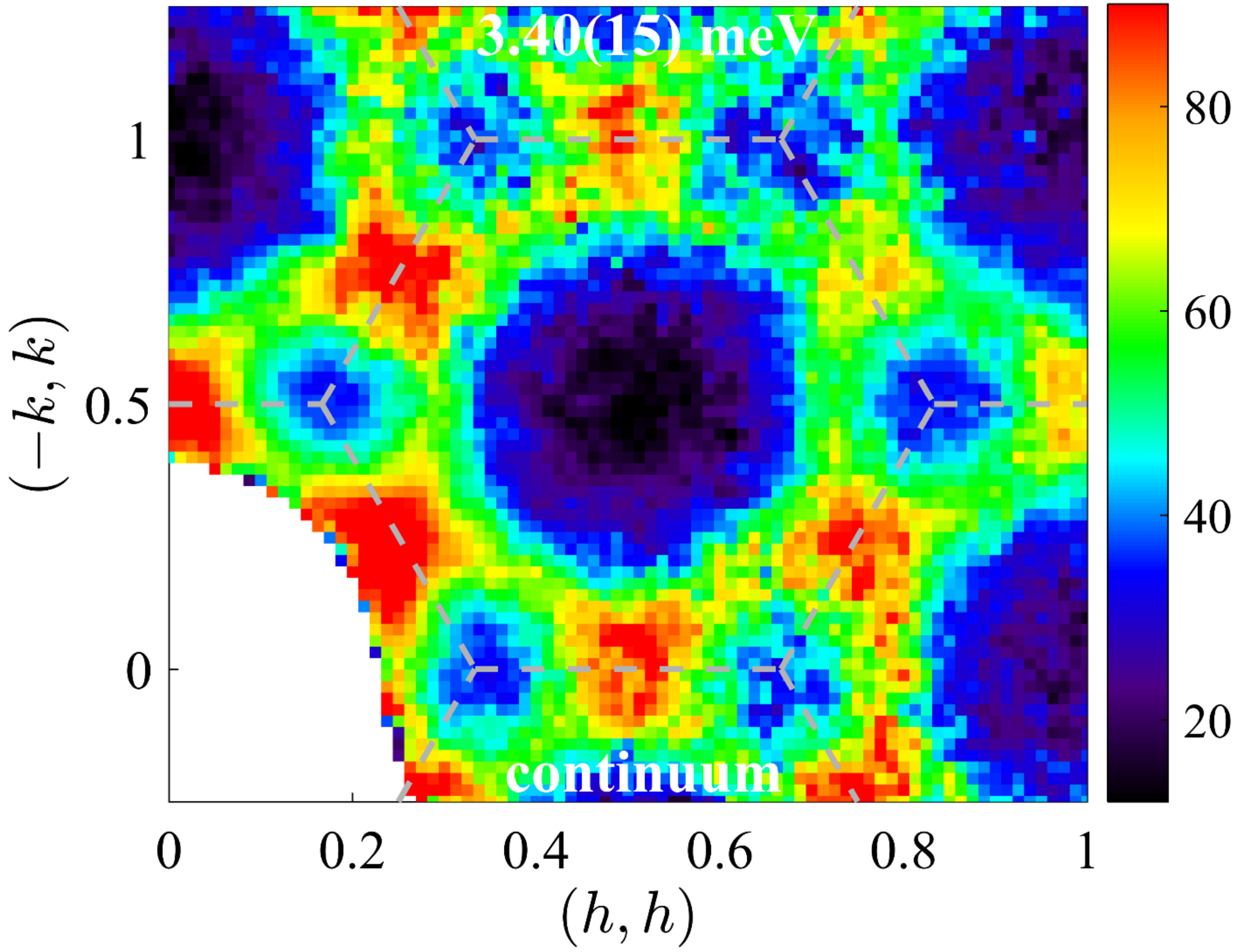
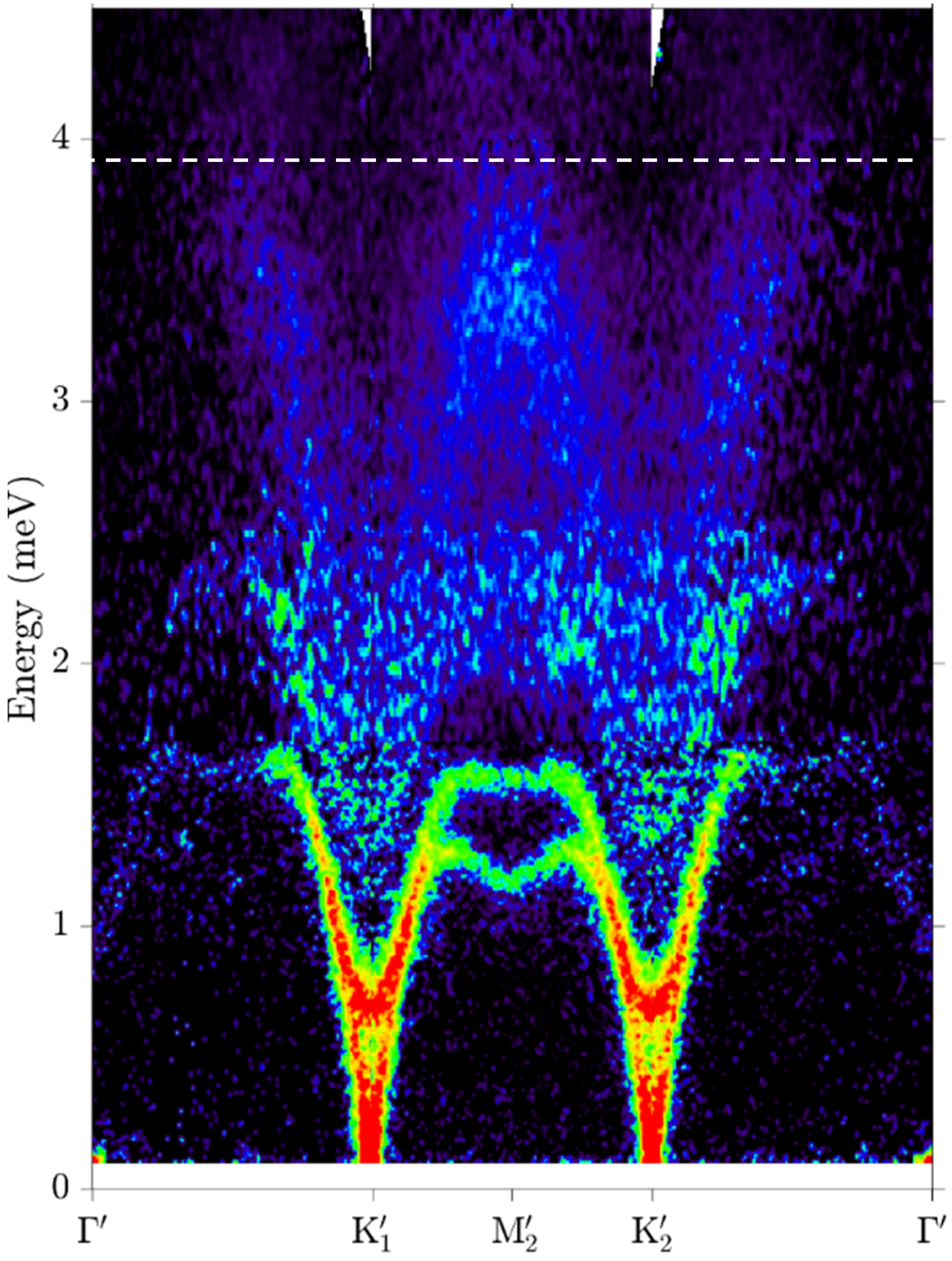
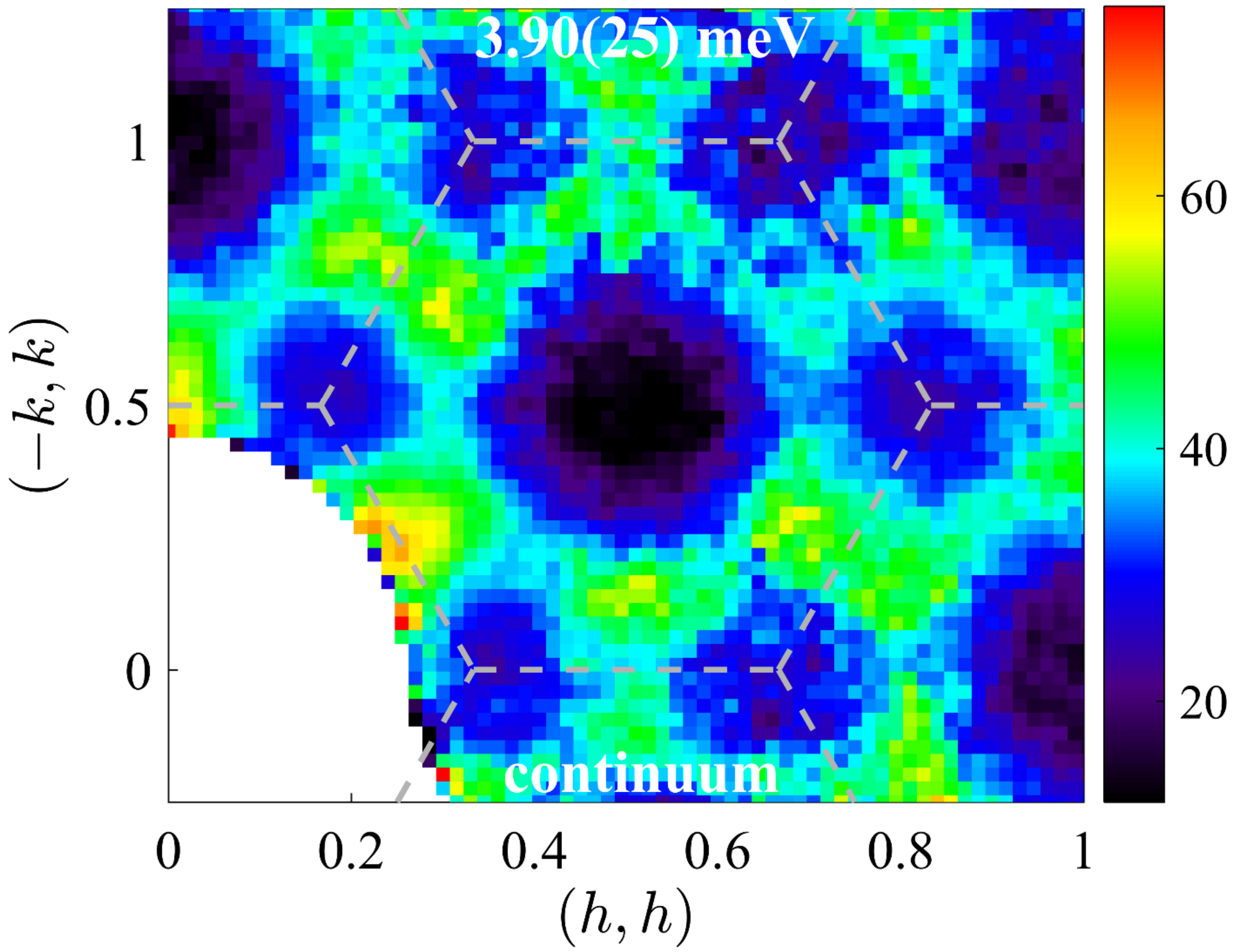


# Intensity modulations in the continuum scattering

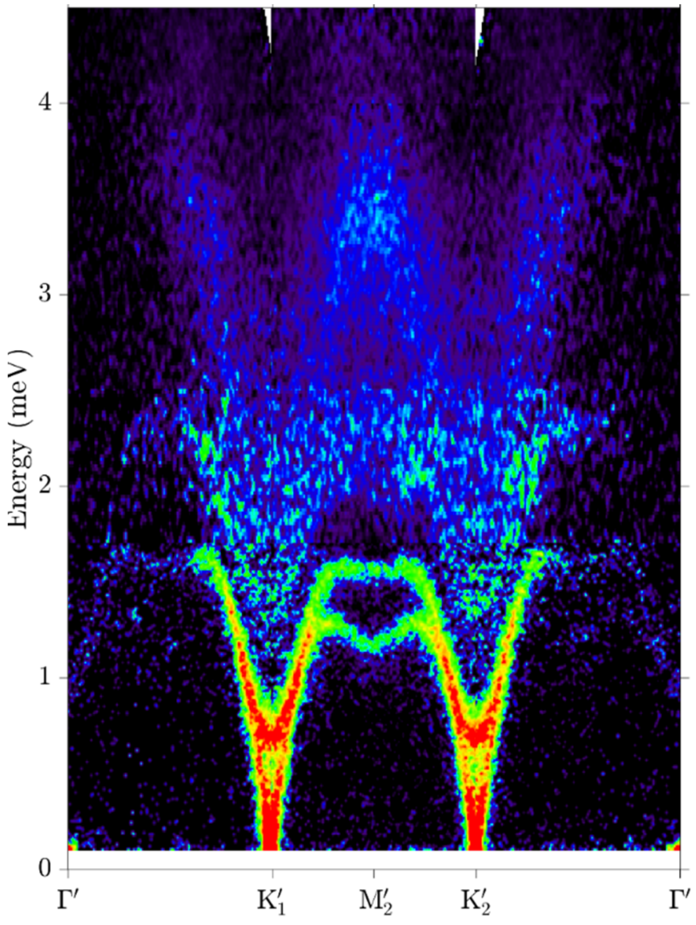
- continuum intensity is highly structured with patterns of rings, triangles and hexagons appearing at various energies

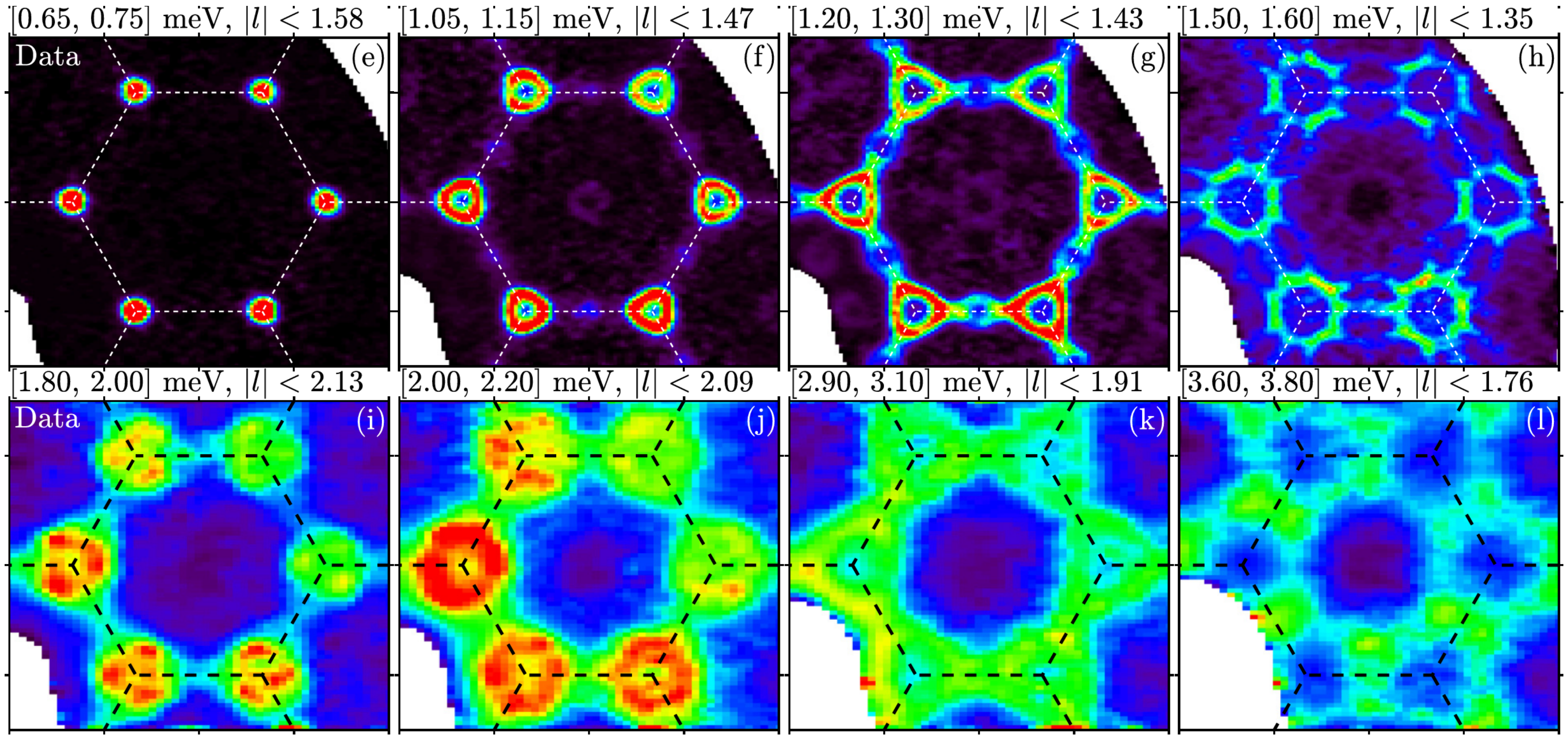


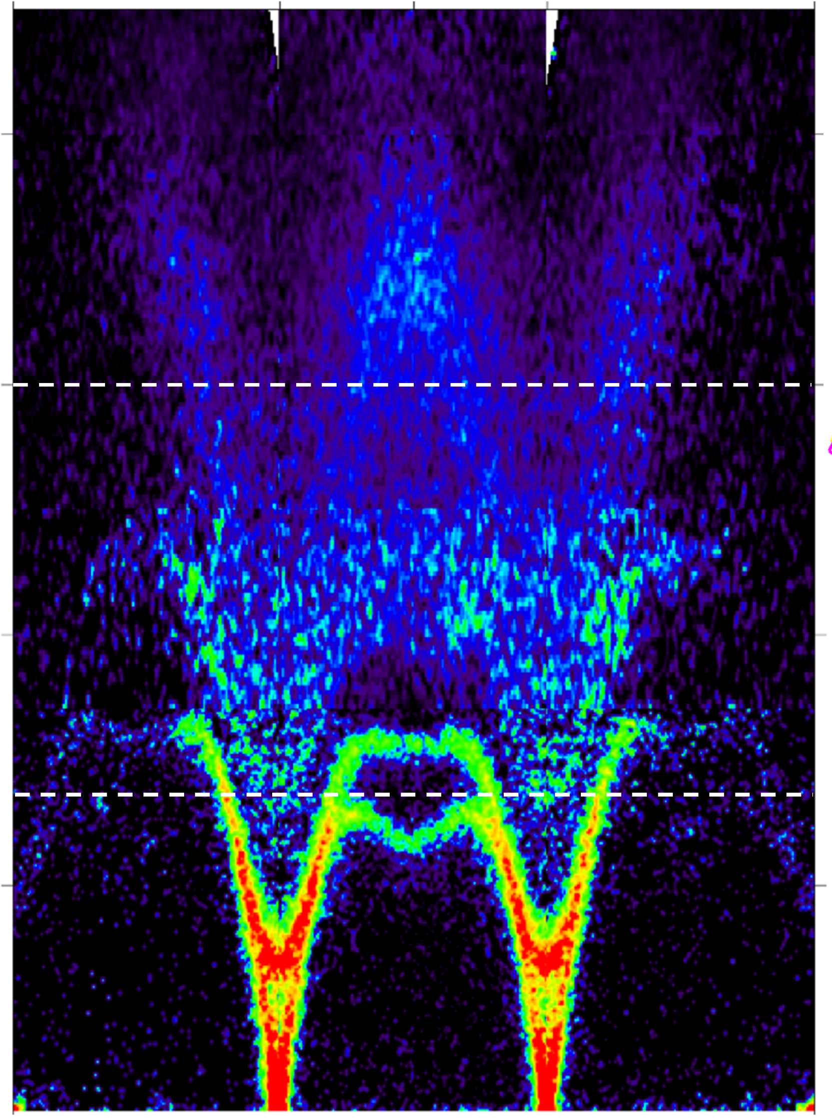
# Intensity modulations in the continuum scattering

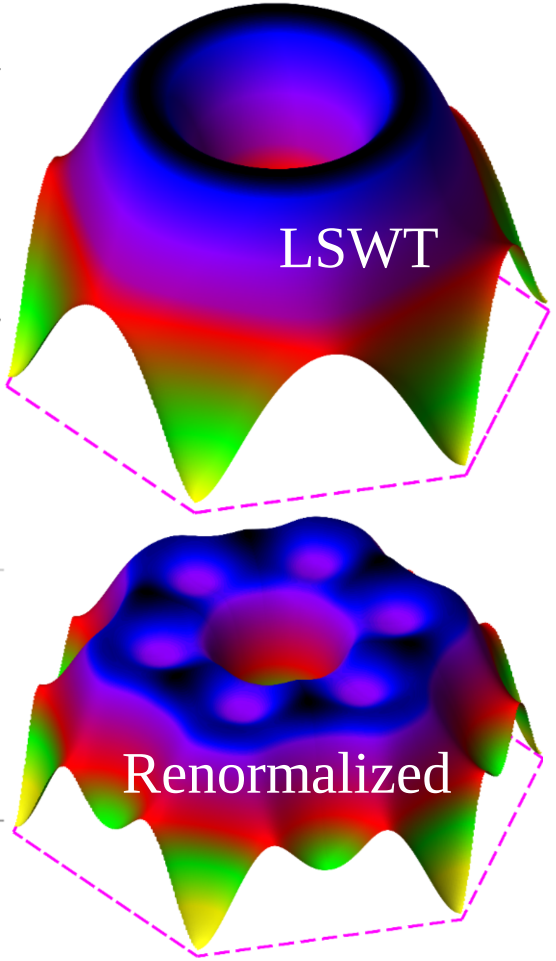
- continuum intensity is highly structured in momentum with patterns of rings, triangles and hexagons appearing at various energies

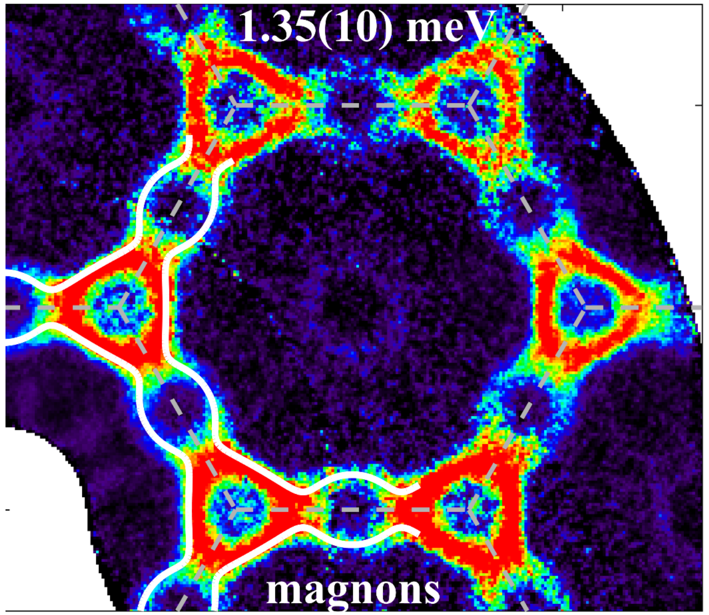


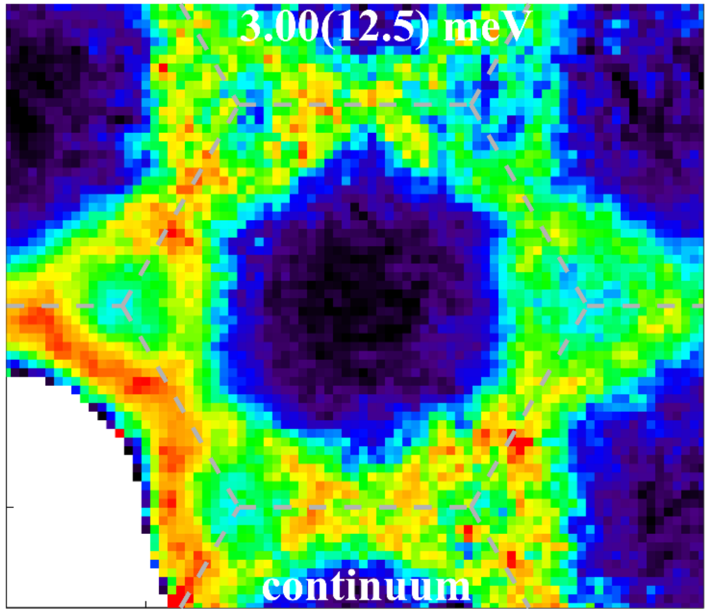



# Summary






- observed sharp magnons throughout, no decay, attribute to strong interaction with continuum
- for the parameterized dispersion no overlap of one and two magnon phase spaces occurs
- magnons carry little weight -> strong transfer of spectral weight to continuum, highly structured continuum intensity, not explained by a two-magnon cross-section