Superspace magnetic structure and topological charges in Weyl semimetal CeAlGe

P. Puphal, et al, submitted (2019)

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Weyl semimetals

Dirac equation (Dirac, 1928).: relativistic fermion S=½ with mass m, invariant to space inversion -1

$$(\widehat{p}_0 + \widehat{\mathbf{p}}\boldsymbol{\sigma})\eta = m\xi, \quad (\widehat{p}_0 - \widehat{\mathbf{p}}\boldsymbol{\sigma})\xi = m\eta.$$

$$\xi = \begin{pmatrix} \xi^1 \\ \xi^2 \end{pmatrix}, \qquad \eta = \begin{pmatrix} \eta_{\dot{1}} \\ \eta_{\dot{2}} \end{pmatrix} \begin{matrix} 1/2 \\ -1/2 \end{matrix}$$

DOI: 10.1103/RevModPhys.90.015001

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mathematician H. Weyl (1929) proposed a simplified version that described massless fermions with a definite chirality (or handedness).

$$(\widehat{p}_0 + \widehat{\mathbf{p}}\boldsymbol{\sigma})\eta = 0.$$

$$(\widehat{p}_0 - \widehat{\mathbf{p}}\boldsymbol{\sigma})\xi = 0.$$

$$E_\pm(p) = \pm p$$

currently no fundamental particles being massless Weyl fermions found

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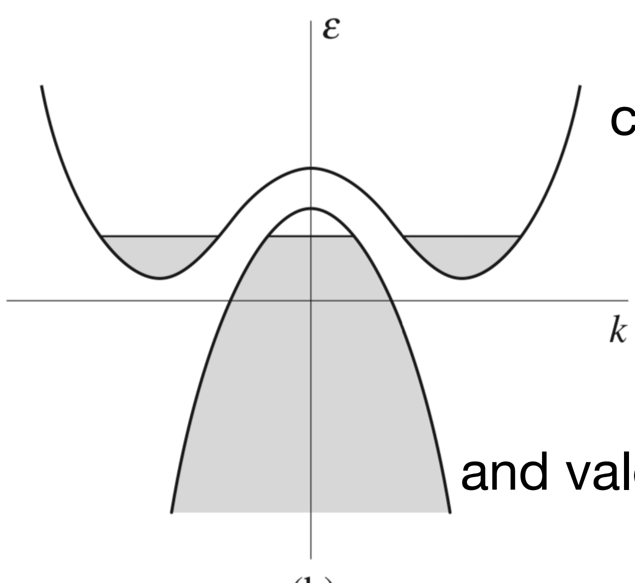
<u>In Condensed Matter Physics</u>

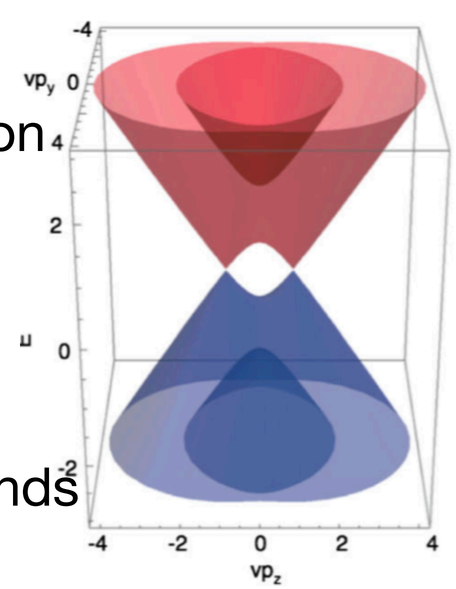
The name “Weyl semimetal” (WSM) was introduced to describe a phase where the chemical potential is near the Weyl nodes. WSM has gapless electronic excitations Weyl fermions that are protected by topology and symmetry.

conventional semi-metal



conduction

and valence bands



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Motivation to study CeAlGe

CeAlGe was predicted theoretically to be an easy-plane FM type-II Weyl semimetal (WSM)*.

It is still not clear if it is WSM… Instead, we have found that CeAlGe is antiferromagnet with rich phase diagram

It has topologically nontrivial magnetization textures in real-space ==> topological Hall effect (THE).

* G. Chang, B. Singh, S.-Y. Xu, G. Bian, S.-M. Huang, C.-H. Hsu, I. Belopolski, N. Alidoust, D. S. Sanchez, H. Zheng, et al., Physical Review B 97 (2018).

Experimental

Samples: both powder and single crystals of CeAlGe grown at PSI in Solid State Chemistry group

BULK SINGLE-CRYSTAL GROWTH OF THE … PHYSICAL REVIEW MATERIALS **3**, 024204 (2019)

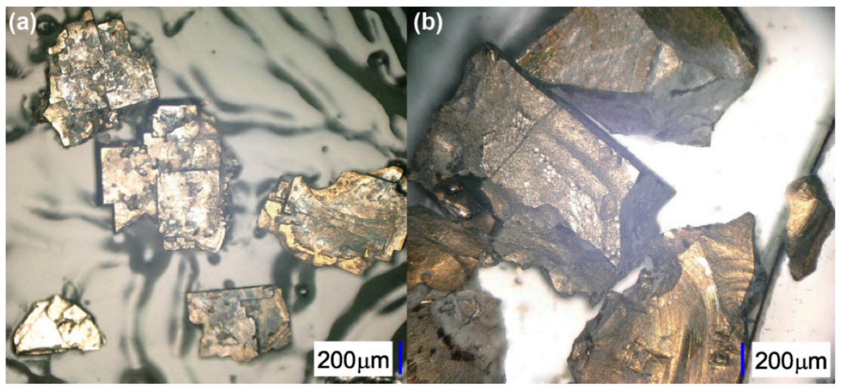


FIG. 2. Pictures of the flux-grown crystals of (a) CeAlGe and (b) PrAlGe right after flux removal using NaOH-H_2O and before su

FIG. 3. Photos of (a) the cast CeAlGe rod, and the floating-zone-own crystals of (b) CeAlGe and (c) PrAlGe.

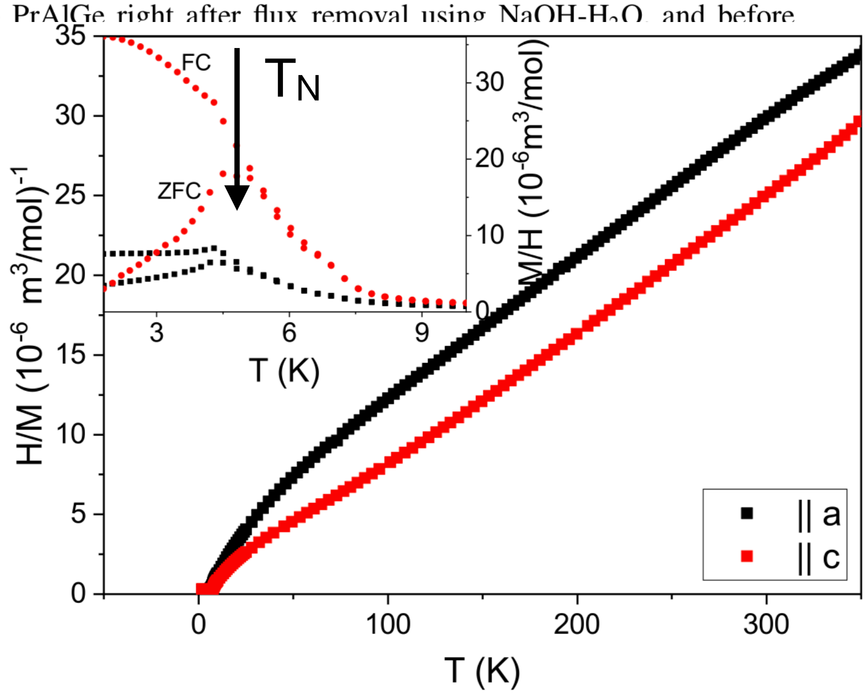
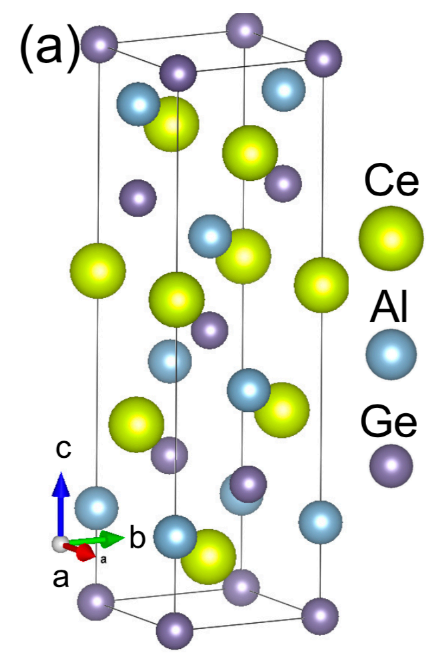


FIG. 8. Magnetic data obtained on a floating-zone-grown CeAlGe single crystal with a mass of 125.4 mg. The magnetic



Space Group: 109 I4_1md C4v-11
non-centrosymmetric
Lattice parameters:
a=4.25717, c=14.64520

Ce1 4a (0,0,z), z=-0.41000 single magnetic Ce site

Neutron diffraction experiments: HRPT and DMC, SANS at PSI Switzerland, D33, at ILL France
Resistivity: Topologicall Hall Effect in University of Tokyo

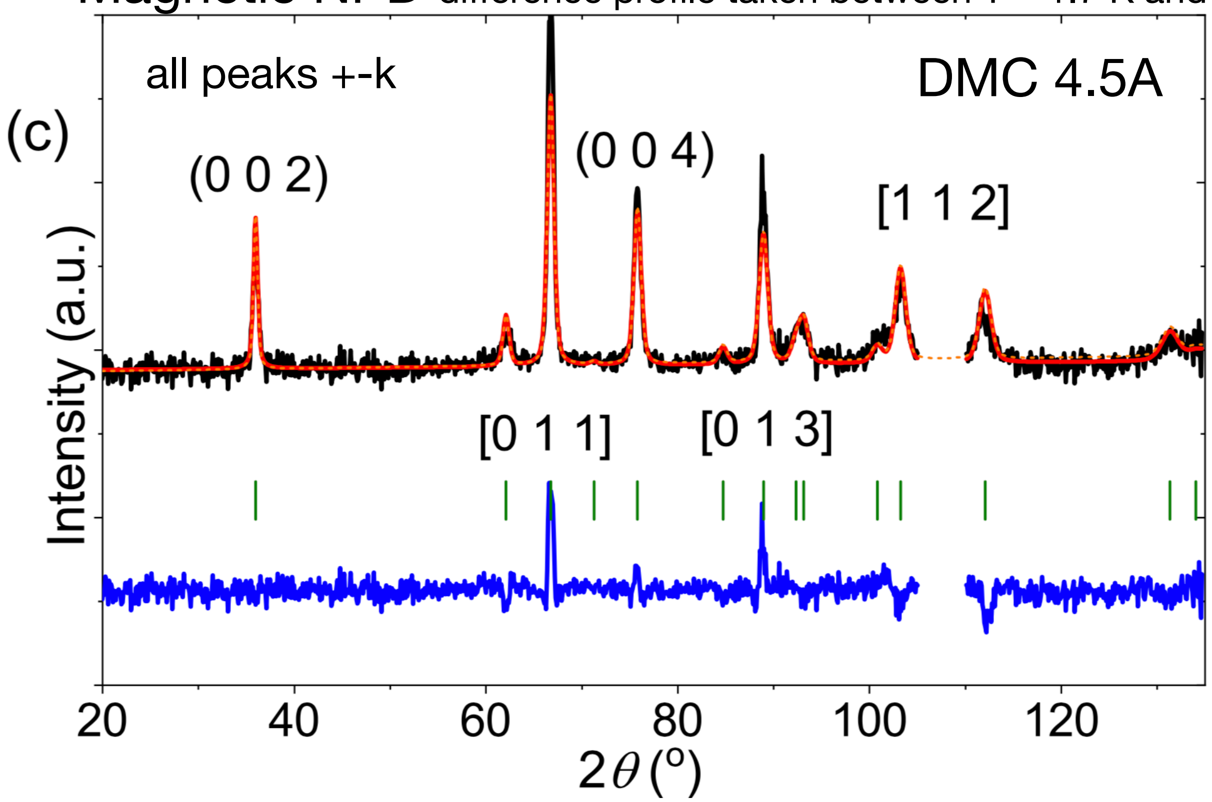
Magnetic peaks are well seen from both powder and s.c. neutron diffraction

CeAlGe

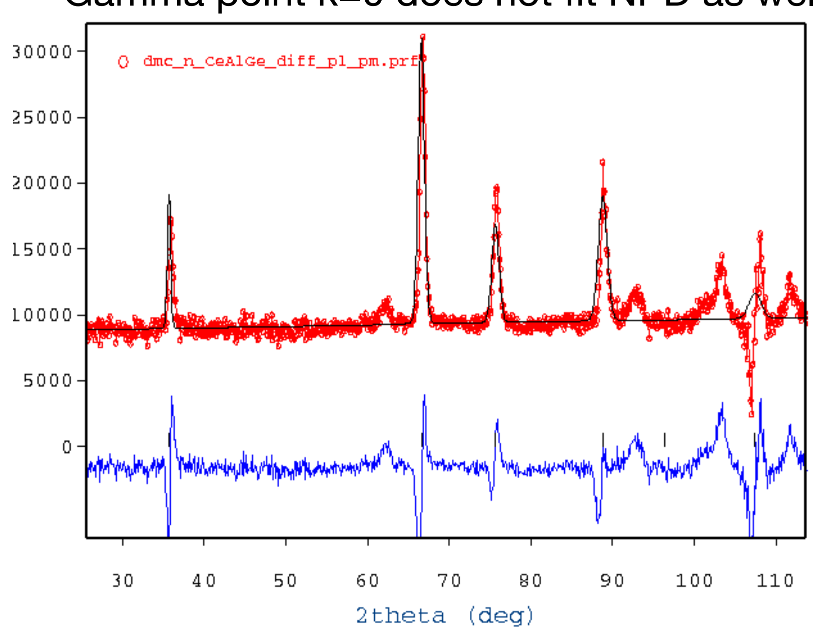
k1=[g,0,0], SM point of BZ, g=0.06503(22) ~65Å

Single crystal

Magnetic NPD difference profile taken between T = 1.7 K and 10 K



Gamma point k=0 does not fit NPD as well



P. Puphal, et al, Physical Review Letters, 124, 017202 (2020)

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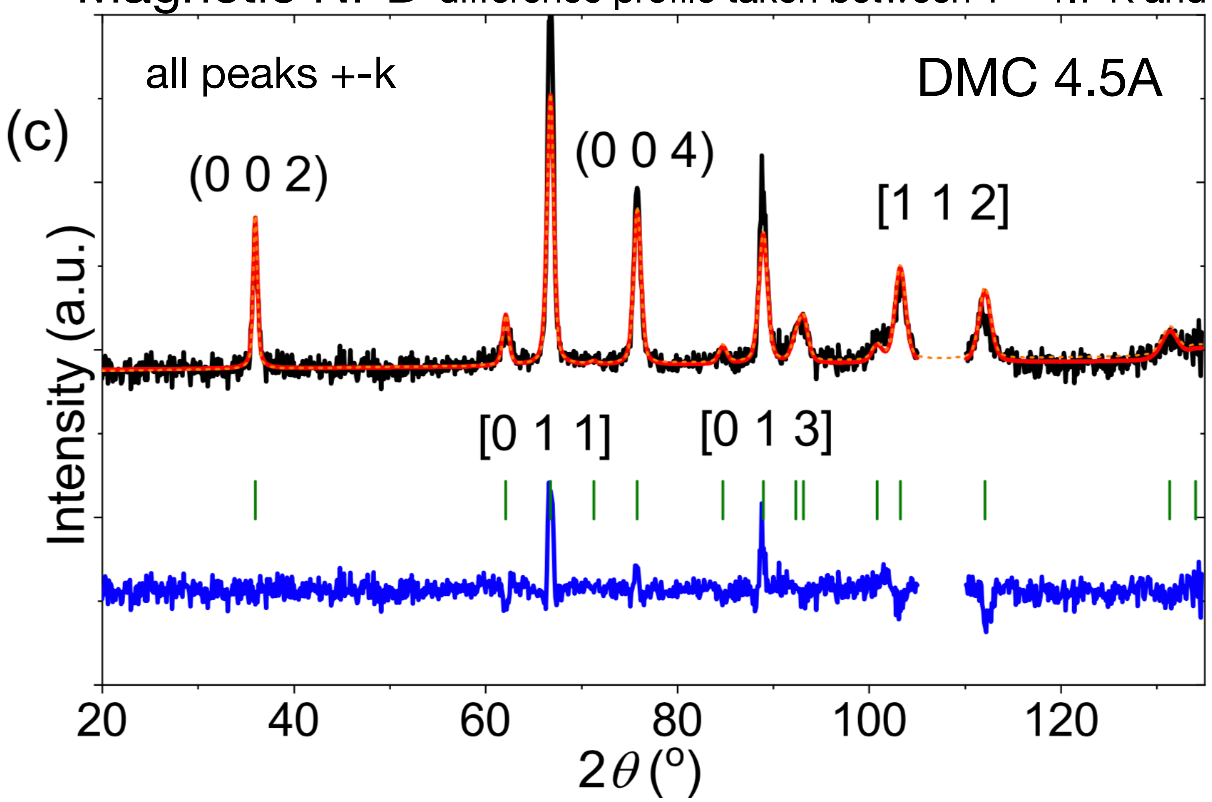
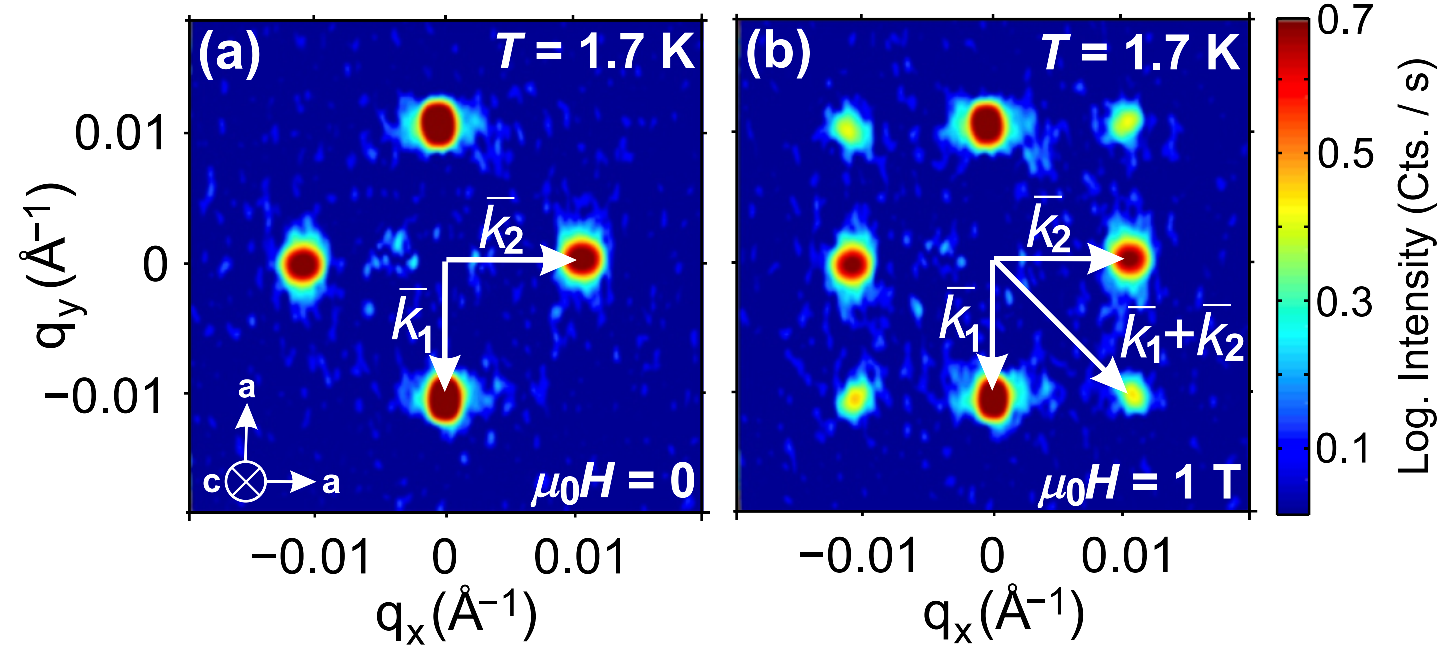


Single crystal

k1=[g,0,0], k2=[0,g,0]



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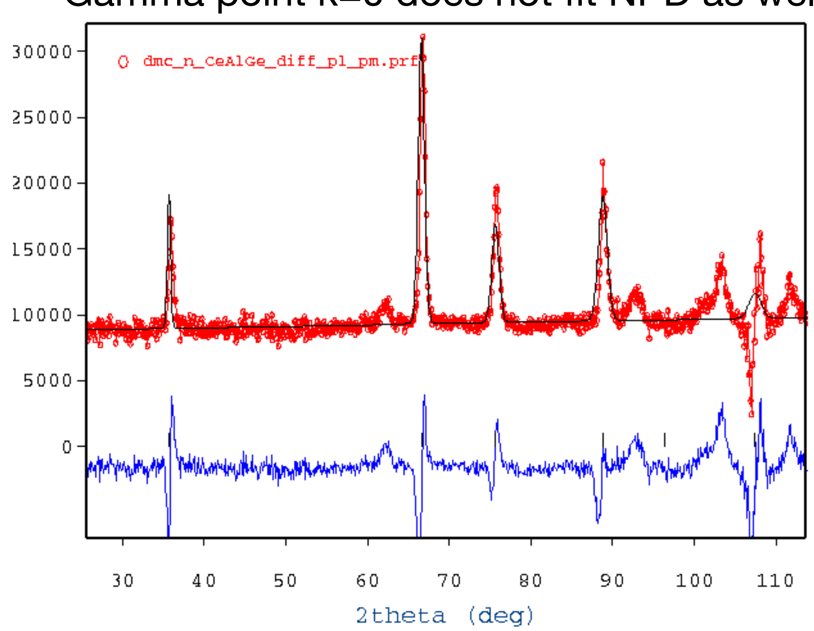

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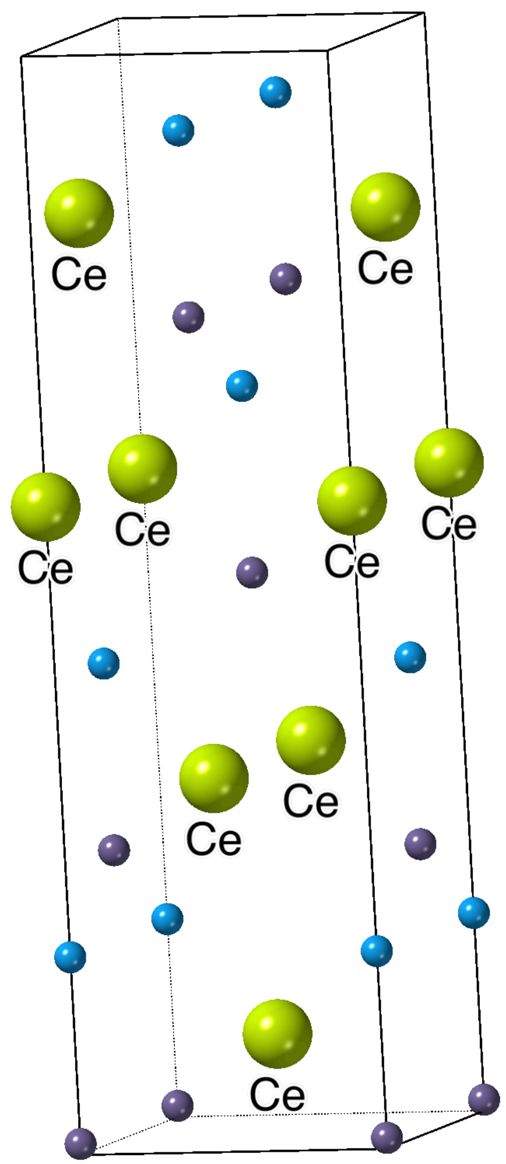
Analysis of magnetic symmetry in CeAlGe

- one propagation vector 1k (±k) magnetic structure
- 2k (full propagation vector star) magnetic structure: actual solution supported by magnetisation, topological hall effect and calculation of topological charges
- both 1k and 2k-structures give similar good description of neutron diffraction intensities

One k-case, standard representation analysis without magnetic group symmetry arguments.

Space group $I4_1md$:
8 symops & I-centering,
Ce 4a (0,0,z) single magnetic Ce site: 4 atoms per cell



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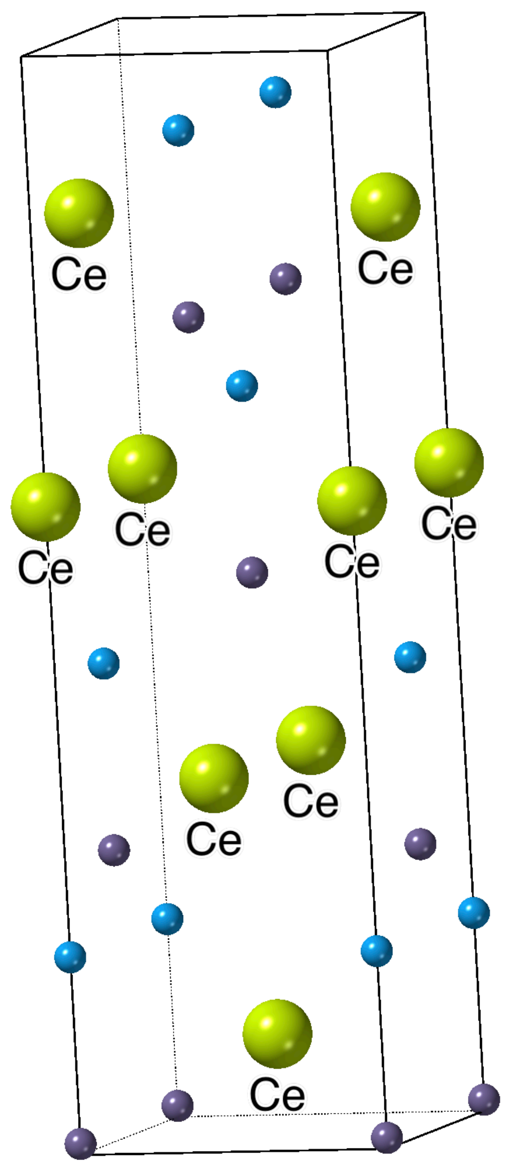
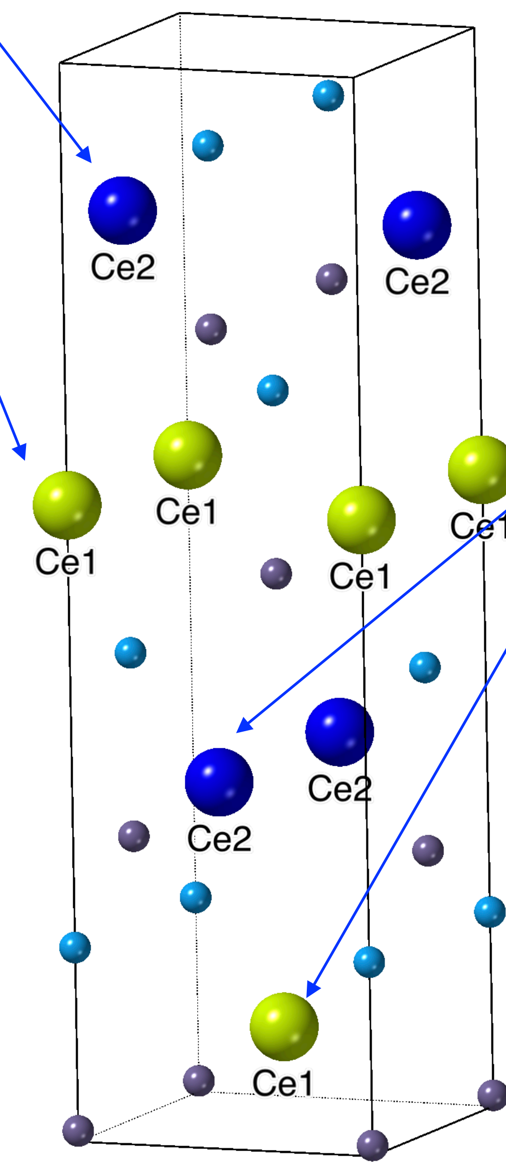
4(a) $\mathrm{Ce1}(0,0,z)$ → $\bar{y}, x+\frac{1}{2}, z+\frac{1}{4}$ → $\mathrm{Ce2}(0,\frac{1}{2},z+\frac{1}{4})$

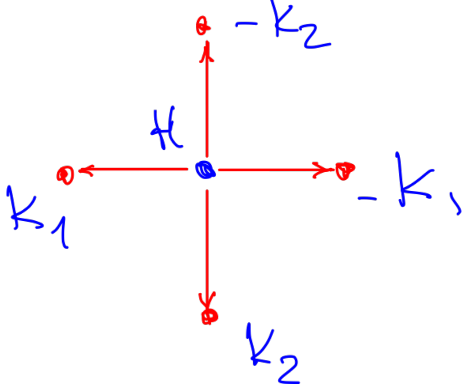




Two other Ce's are generated by I-centering translations (½, ½, ½)+

One k-case, standard representation analysis without magnetic group symmetry arguments: Space group $I4_1md$, Ce 4a (0,0,z)

Propagation vector star $\{[g,0,0],[0,g,0]\}$



$\boldsymbol{k}_1 \rightarrow \boldsymbol{k}_2$

$$\bar{y}, x+\frac{1}{2}, z+\frac{1}{4}$$

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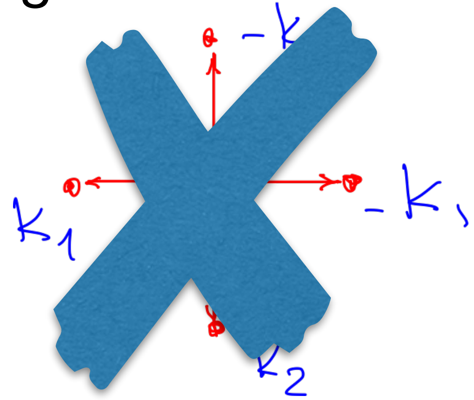

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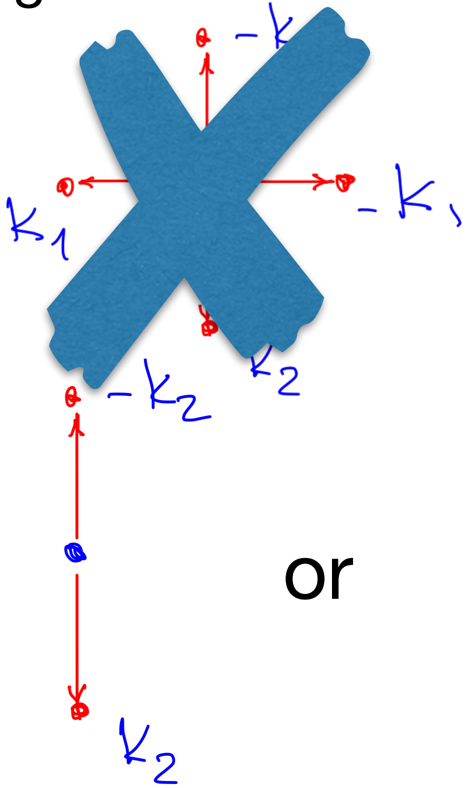
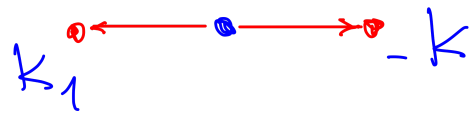


or



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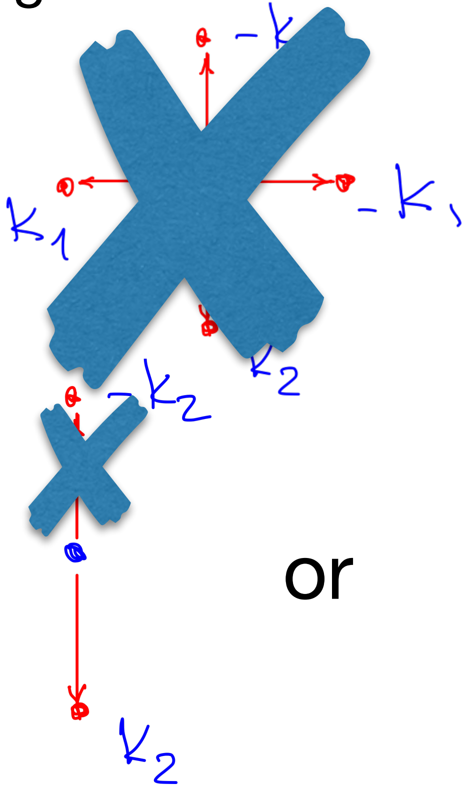

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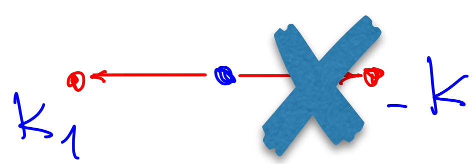

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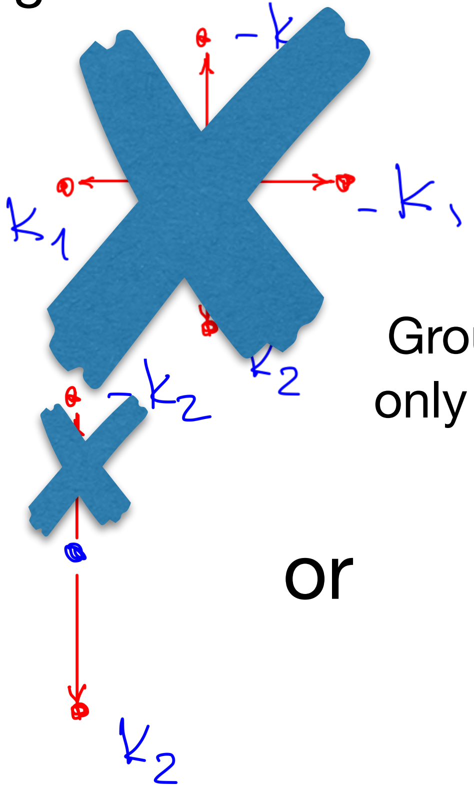
Propagation vector star $\{[g,0,0],[0,g,0]\}$

$\boldsymbol{k}_1 \rightarrow \boldsymbol{k}_2$

$\bar{y}, x+\frac{1}{2}, z+\frac{1}{4}$

Group G_k has x,y,z
only 2 symops $x,\bar{y},z$ out of 8!



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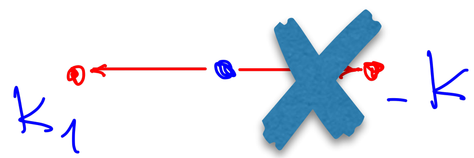

$Ce1(0,0,z)$
$Ce2(0,\frac{1}{2},z+\frac{1}{4})$

Two independent sites. No symmetry relations between Ce1 and Ce2

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$$\mathrm{Ce2}(0,\frac{1}{2},z+\frac{1}{4})$$

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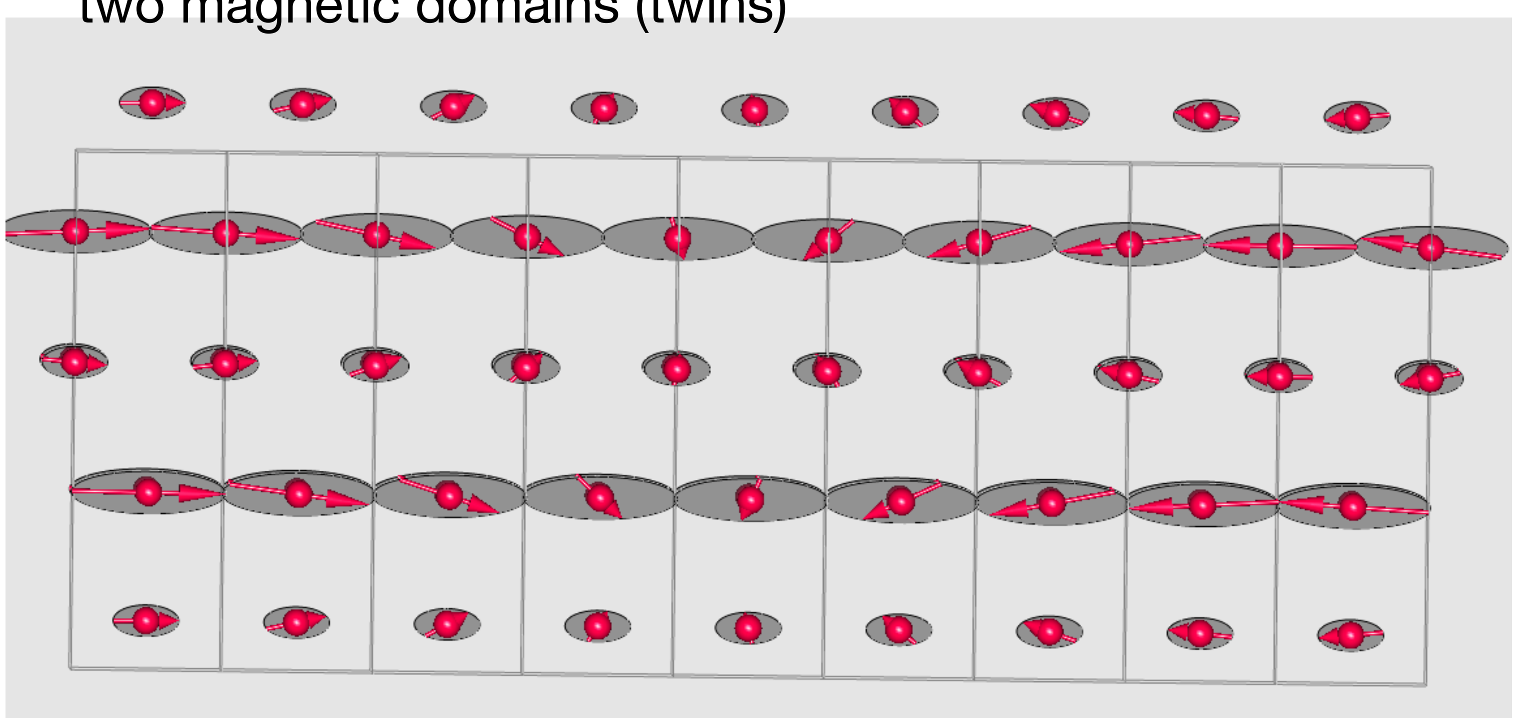
$k=|\boldsymbol{k}_1|=|\boldsymbol{k}_2|=g$

$$\mathbf{M}_{Ce(i)} = m_{ix}\sin(2\pi kx)\mathbf{e}_{\boldsymbol{x}} + m_{iz}\sin(2\pi kx+\varphi_i)\mathbf{e}_{\boldsymbol{z}}, \quad i=1,2$$

Experimental values:
Ce1: m_{1x}= -0.64(1), m_{1z} =-0.30(6)
Ce2: m_{2x}= -1.50(2), m2z = 0.46(8)

$\varphi_1=\varphi_2\approx 90°$

Solution: tau2/SM2 irreducible representation
Cycloid in ac-plane for k1=[g,0,0], in bc=lane for k2=[0,g,0]
two magnetic domains (twins)



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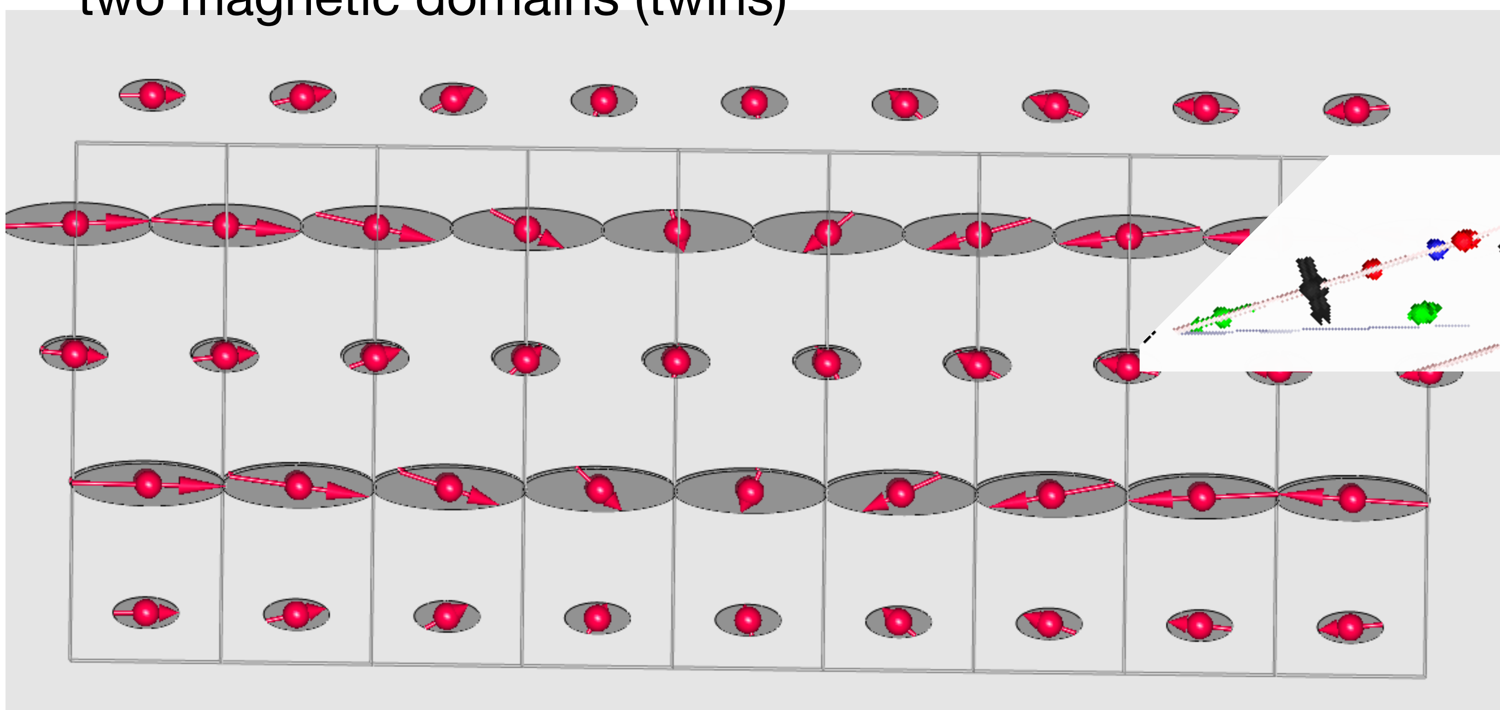

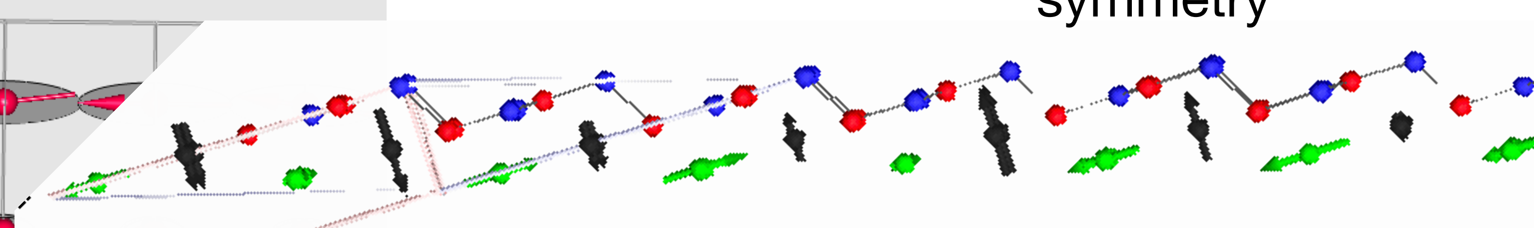

Note: if $\varphi_1=\varphi_2=0$ → amplitude modulation, different symmetry

Symmetry of cycloid. 3D+1 superspace group for SM2 irrep

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Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

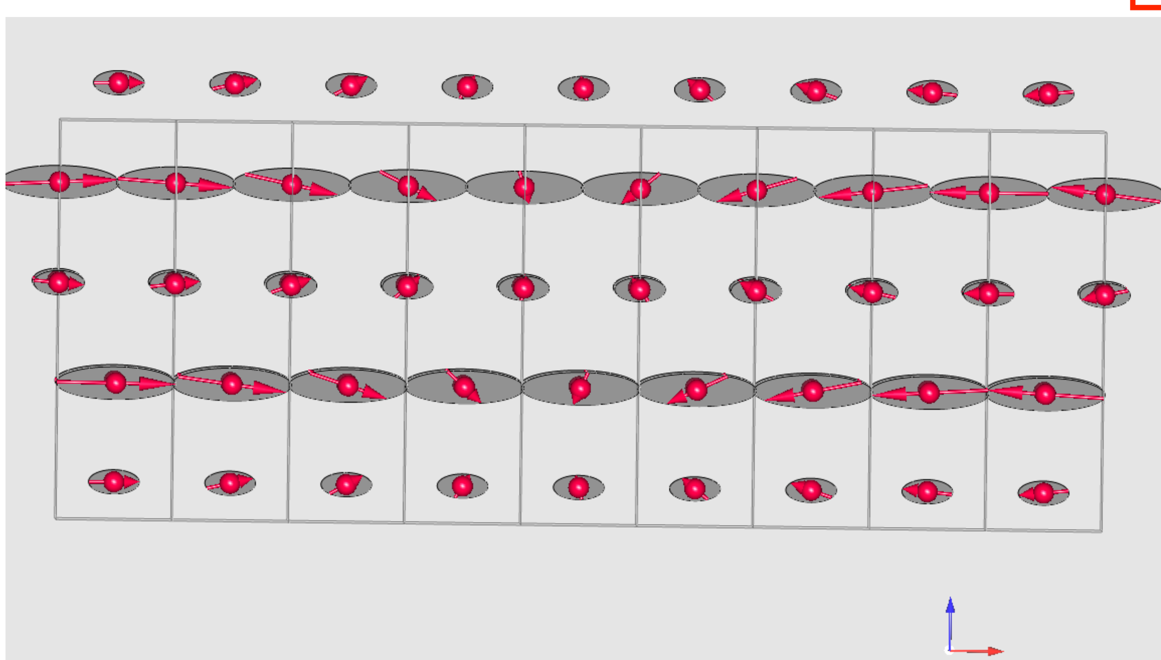
Advantage of magnetic symmetry even for 1k-case

$I4_1md1'$

↓

I2mm1'(0,0,g)0s0s, basis={(0,0,-1,0),(0,1,0,0),(1,0,0,0),(0,0,0,1)}, k-active= (g,0,0)

atom	site	x	y	z	occ	mx	my	mz	
Ce1_1	2a	0.41000	0.00000	0.00000	1.00000				
						mx1	0	mz1	k1 amplitude
						0	0.00000	90	k1 phase, degrees
Ce1_2	2b	0.66000	0.00000	0.50000	1.00000				
						mx2	0	mz2	k1 amplitude
						0	0.00000	90	k1 phase, degrees



$$\mathbf{M}_{Ce(i)} = m_{ix}\sin(2\pi kx)\mathbf{e}_{\boldsymbol{x}} + m_{iz}\cos(2\pi kx)\mathbf{e}_{\boldsymbol{z}}, \quad i = 1, 2$$

phase shift 90 degrees is fixed by symmetry!

Experimental values:

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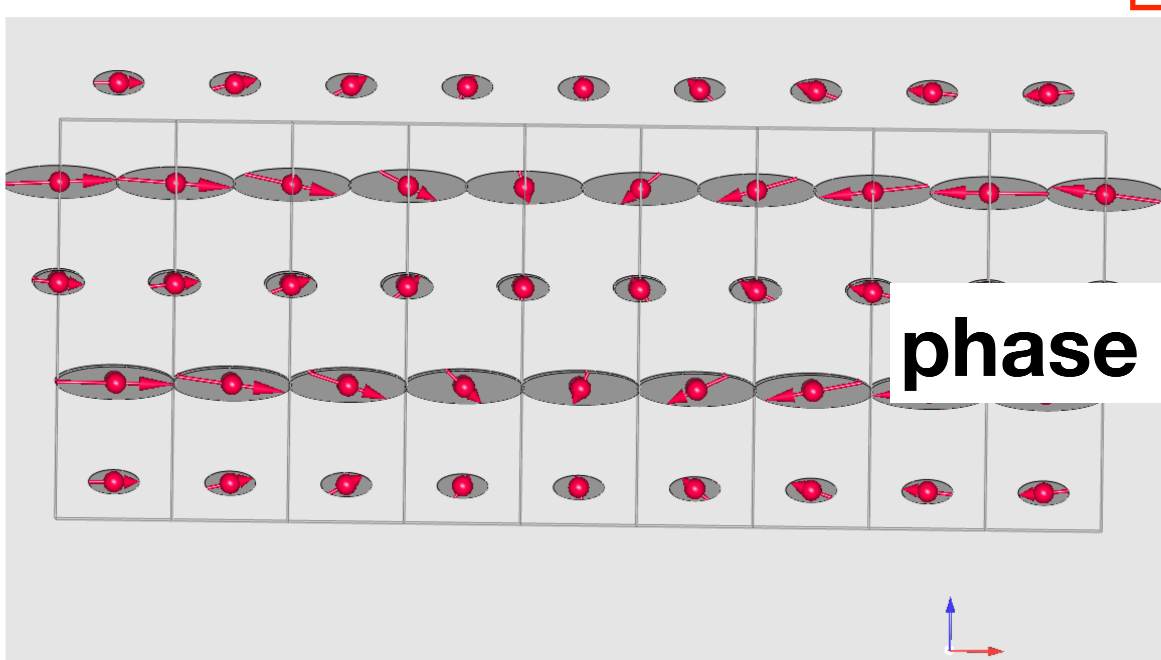
						0	0.00000	90	k1 phase, degrees



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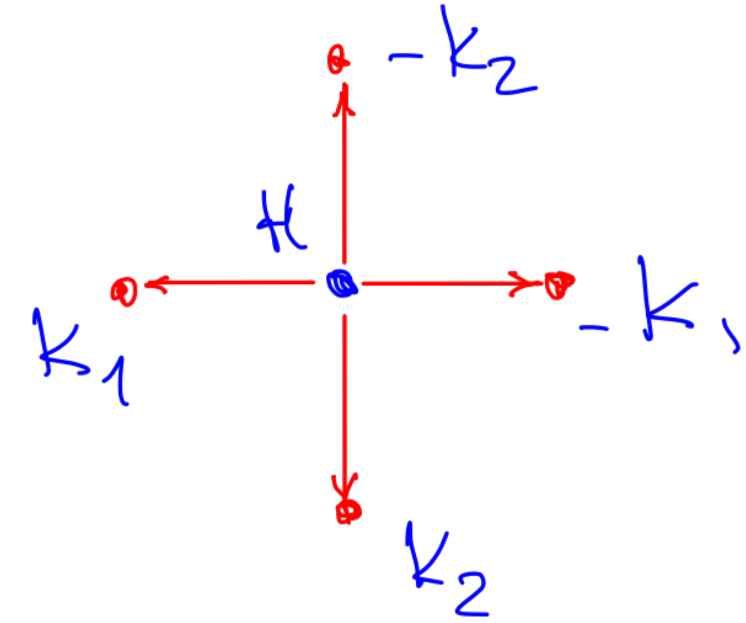
CeAlGe: Maximal symmetry full star superspace 3D+2 magnetic group I4_1md1'(a00)000s(0a0)0s0s

Parent Space Group: 109 I4_1md C4v-11,
Ce1 4a (0,0,z), z=-0.41000 single Ce site

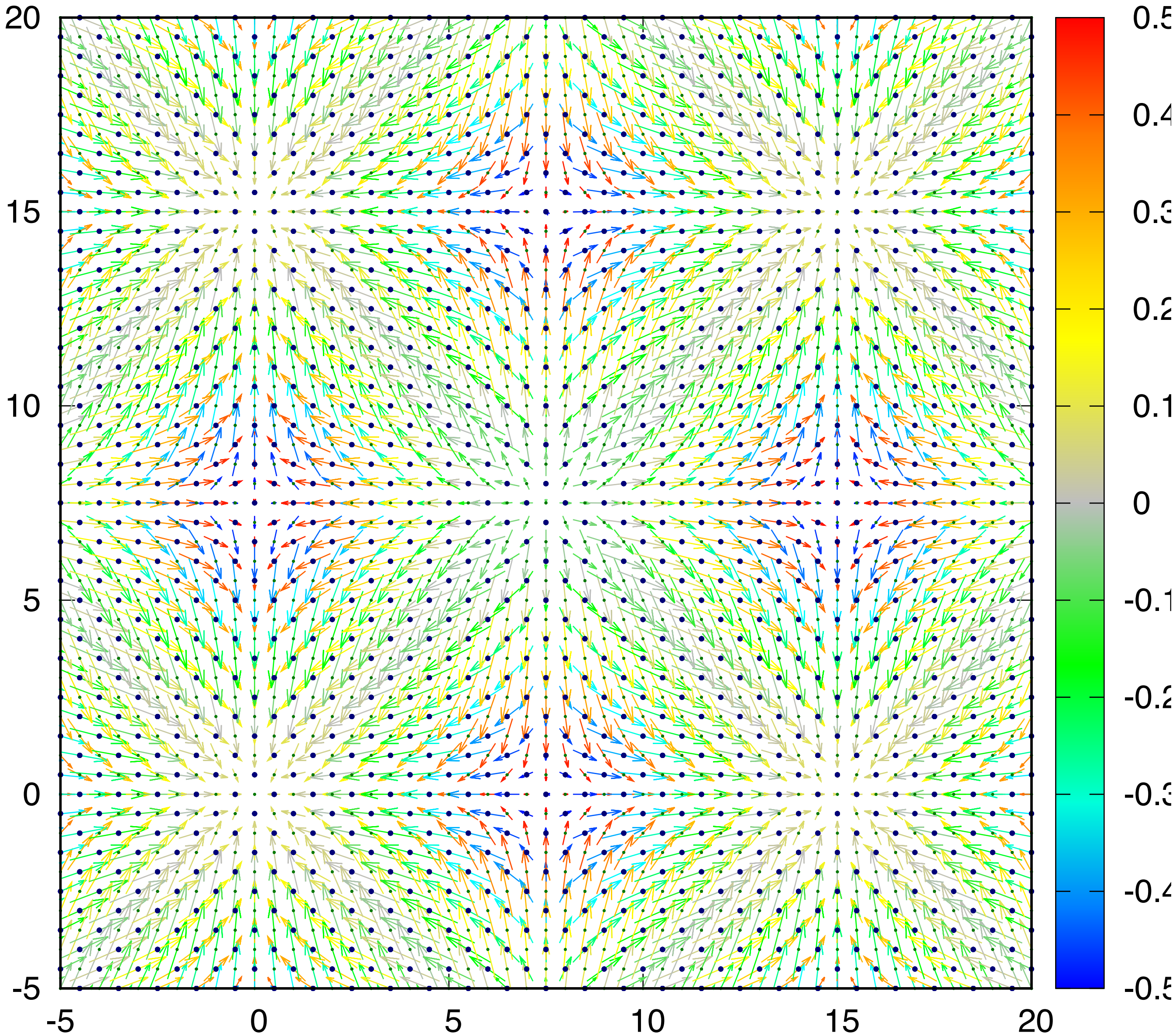
IR: mSM2 , k-active= (g,0,0),(0,g,0)
P (g,0;g,0) 109.2.67.4.m240.? I4_1md1'(a,0,0)000s(0,a,0)0s0s

k1=[g,0,0], SM point of BZ,
g=0.06503(22): four arms



View along the z-(c-)axis of the magnetic structure of CeAlGe.
The x- and y-axes are in units of in-plane lattice parameter a.

(M_x,M_y) components in the xy plane, M_z-component by color



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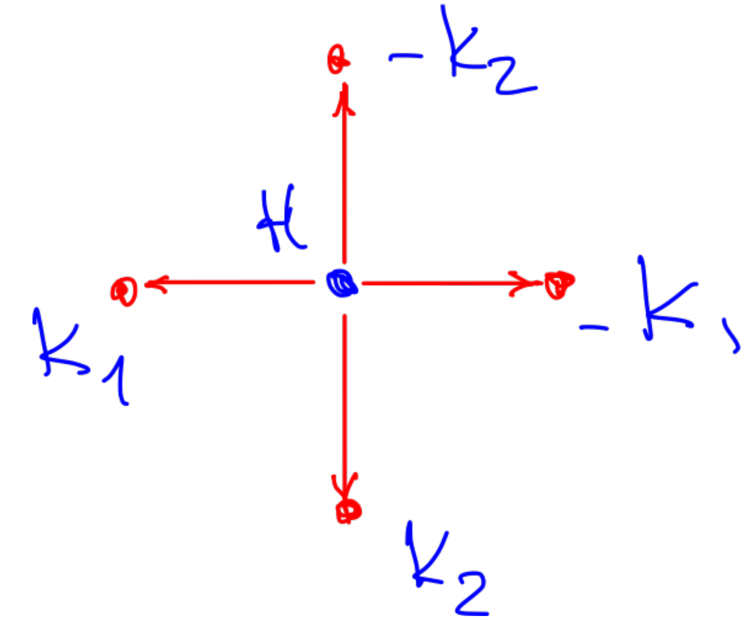

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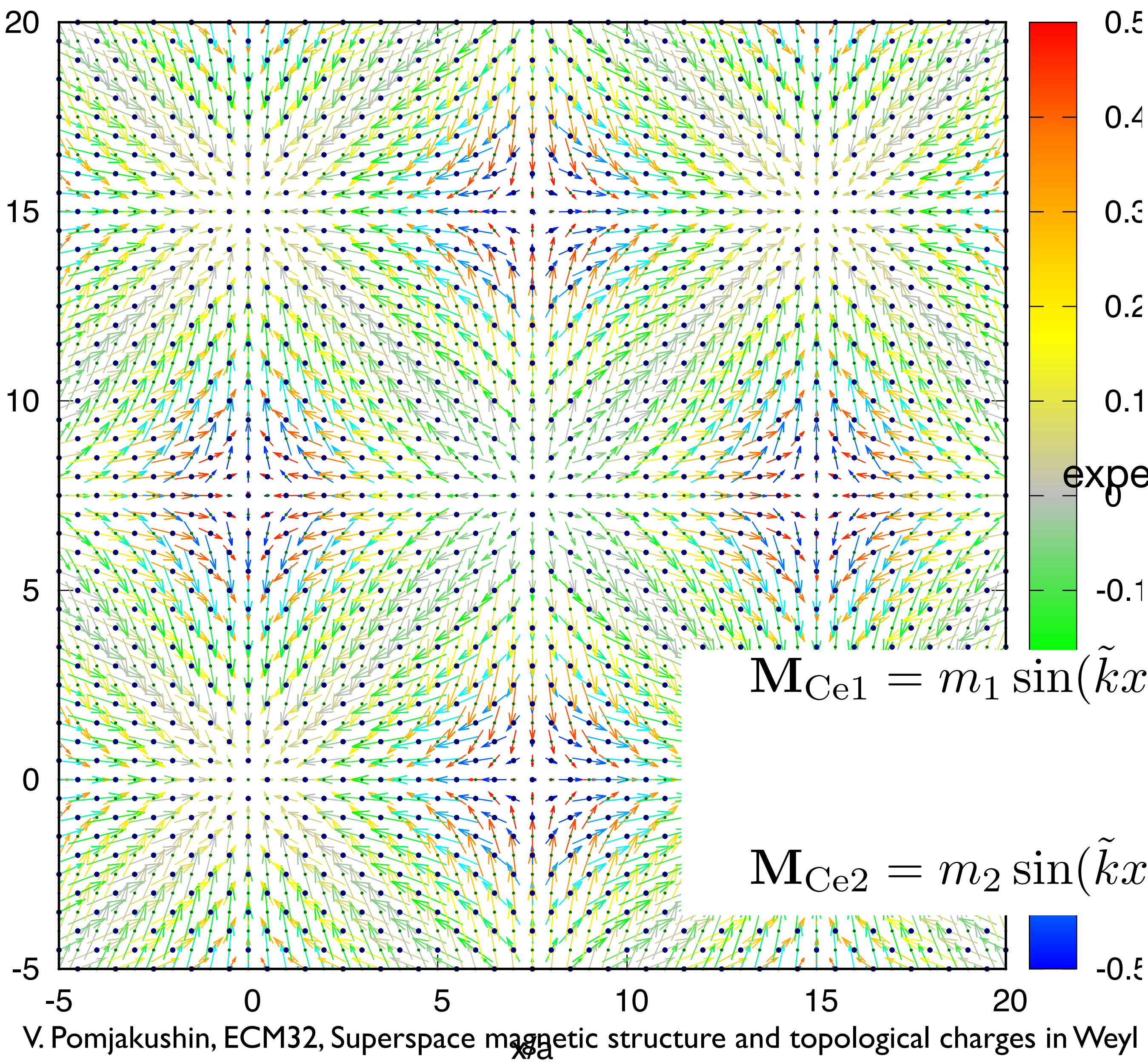


All Ce are equivalent and their moments are given symmetrically by 4 parameters

experiment: (m1,m2,m3,m4) = (0.44(1), 1.02(1), −0.21(5), 0.29(7)) μB.

$\tilde{k}=2\pi|\mathbf{k}_1|=2\pi|\mathbf{k}_2|=2\pi g$

$$\mathbf{M}_{\mathrm{Ce1}} = m_1 \sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_2 \sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \Big(m_3 \cos(\tilde{k}x) + m_4 \cos(\tilde{k}y)\Big)\,\mathbf{e}_{\boldsymbol{z}}$$

$$\mathbf{M}_{\mathrm{Ce2}} = m_2 \sin(\tilde{k}x)\mathbf{e}_{\boldsymbol{x}} + m_1 \sin(\tilde{k}y)\mathbf{e}_{\boldsymbol{y}} + \Big(m_4 \cos(\tilde{k}x) + m_3 \cos(\tilde{k}y)\Big)\,\mathbf{e}_{\boldsymbol{z}}$$

ISOTROPY Software Suite

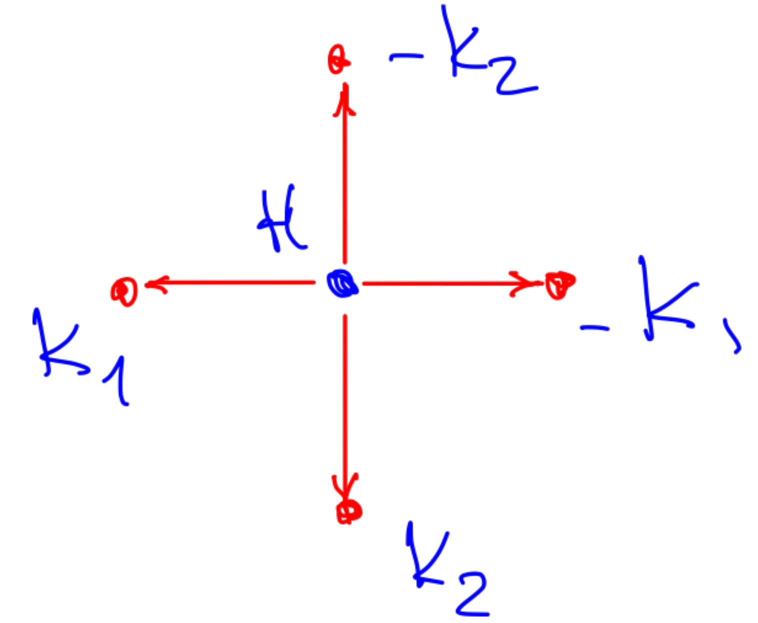
Harold T. Stokes, Dorian M. Hatch, and Branton J. Campbell, Department of Physics and Astronomy, Brigham Young University, Provo, Utah 84606, USA,

CeAlGe: Maximal symmetry full star superspace 3D+2 magnetic group I4_1md1'(a00)000s(0a0)0s0s

Parent Space Group: 109 I4_1md C4v-11,
Ce1 4a (0,0,z), z=-0.41000 single Ce site

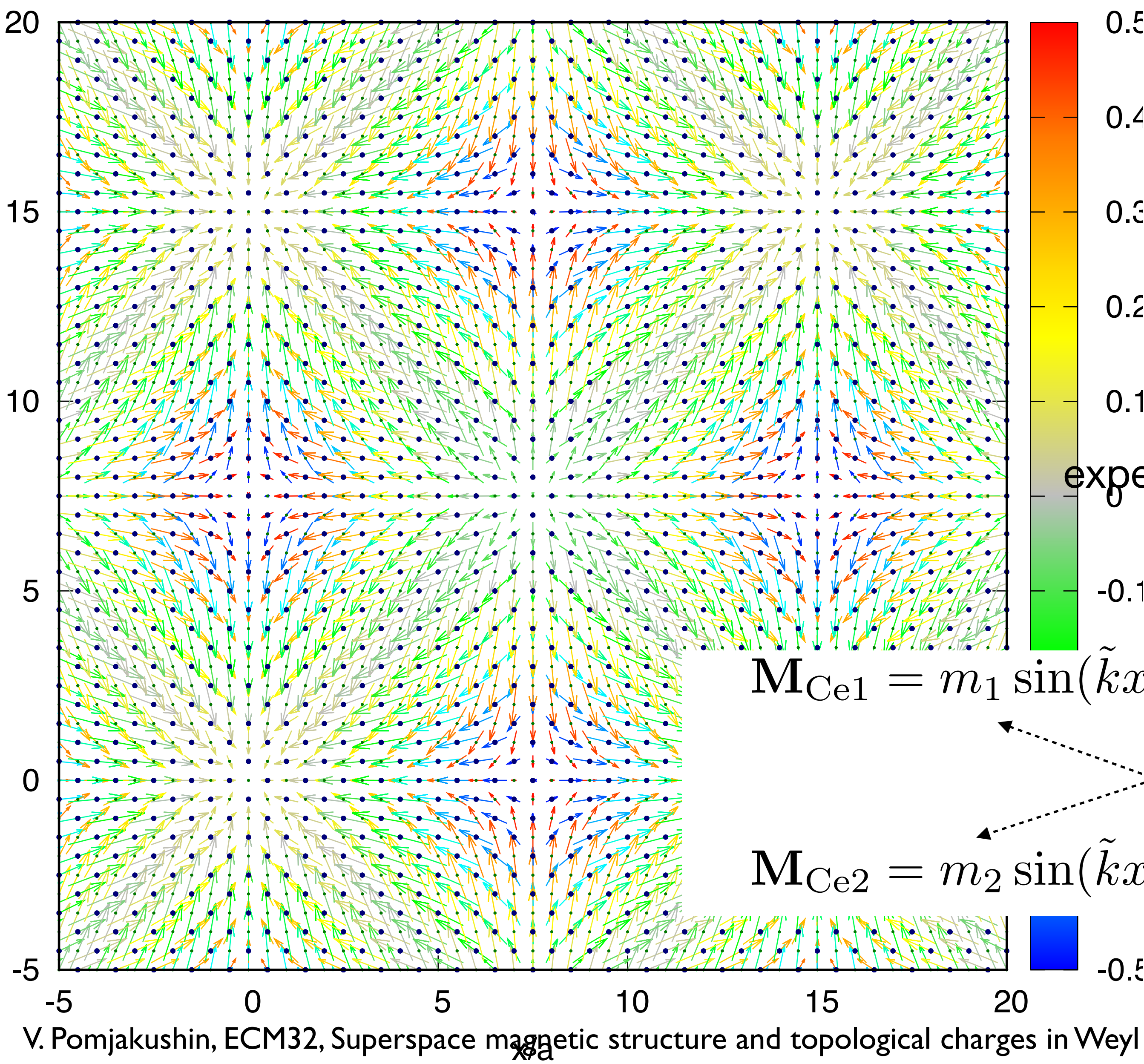
IR: mSM2 , k-active= (g,0,0),(0,g,0)
P (g,0;g,0) 109.2.67.4.m240.? I4_1md1'(a,0,0)000s(0,a,0)0s0s

k1=[g,0,0], SM point of BZ,
g=0.06503(22): four arms



View along the z-(c-)axis of the magnetic structure of CeAlGe.
The x- and y-axes are in units of in-plane lattice parameter a.

(M_x,M_y) components in the xy plane, M_z-component by color



All Ce are equivalent and their moments are given symmetrically by 4 parameters

experiment: (m1,m2,m3,m4) = (0.44(1), 1.02(1), −0.21(5), 0.29(7)) μ_B.

$\tilde{k}=2\pi|\mathbf{k}_1|=2\pi|\mathbf{k}_2|=2\pi g$

$$\mathbf{M}_{\mathrm{Ce1}} = m_1 \sin(\tilde{k}x)\mathbf{e}_x + m_2 \sin(\tilde{k}y)\mathbf{e}_y + \left(m_3 \cos(\tilde{k}x) + m_4 \cos(\tilde{k}y)\right)\mathbf{e}_z$$

$$\mathbf{M}_{\mathrm{Ce2}} = m_2 \sin(\tilde{k}x)\mathbf{e}_x + m_1 \sin(\tilde{k}y)\mathbf{e}_y + \left(m_4 \cos(\tilde{k}x) + m_3 \cos(\tilde{k}y)\right)\mathbf{e}_z$$

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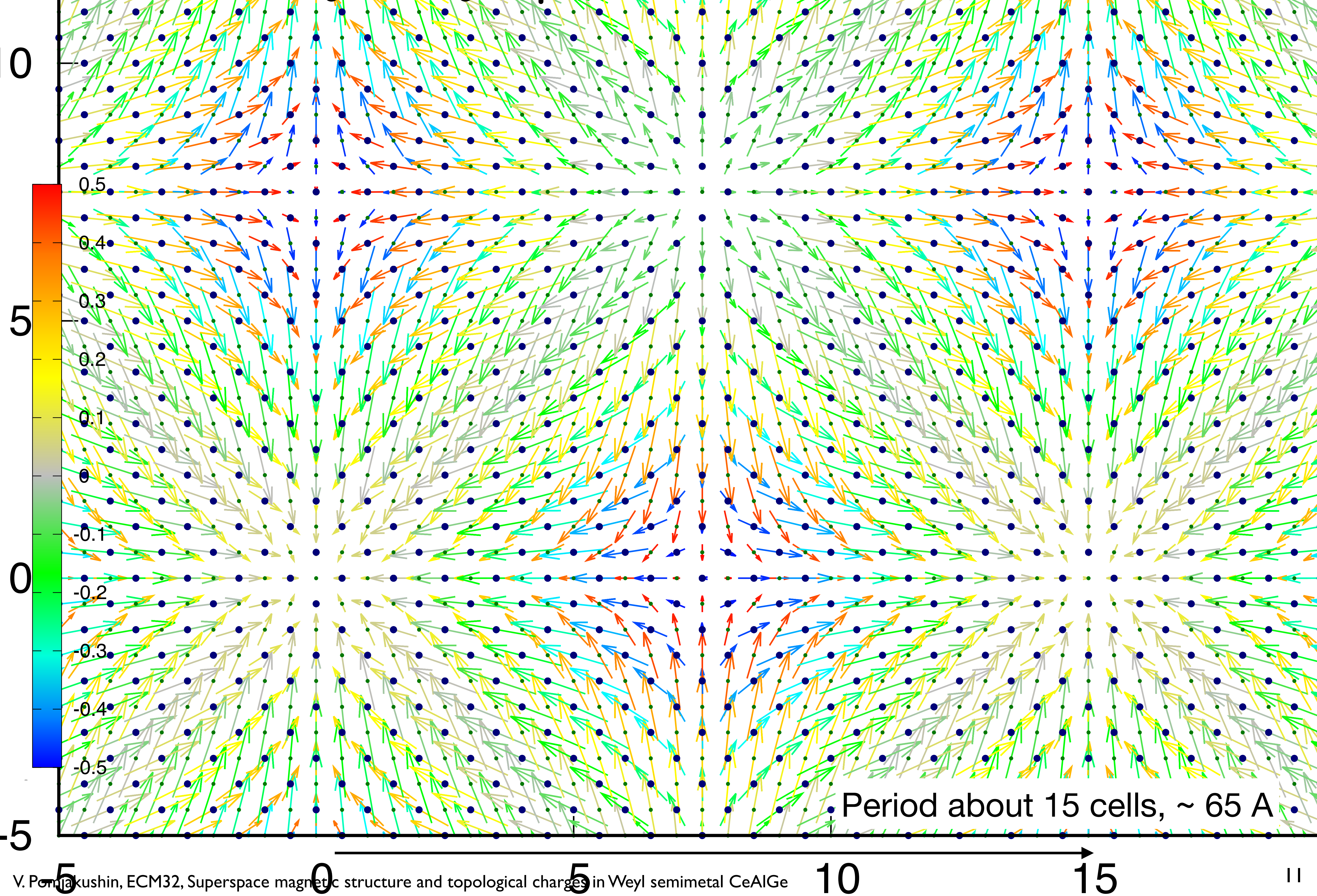
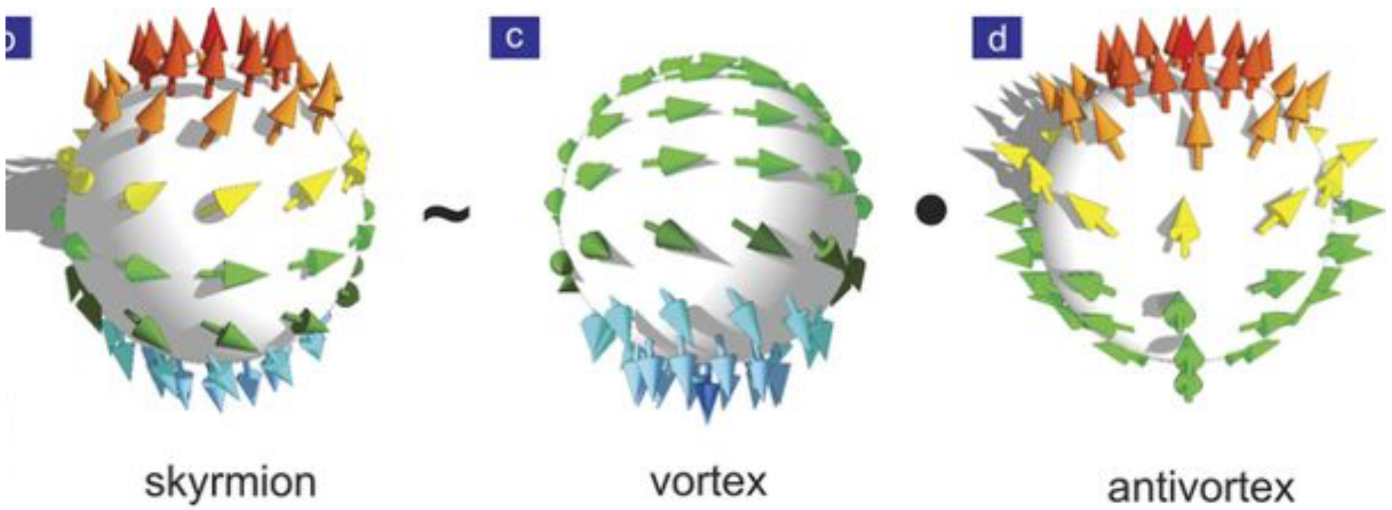


Skyrmion

- T Skyrme was a British physicist. In 1962 he proposed topological soliton to model a particle like neutron or proton. These entities would later in 1982 became known as skyrmions.



Stereographic-projection view of the magnetisation distribution of a single magnetic skyrmion *Scientific Reports **5**, Article number: 15773 (2015)*

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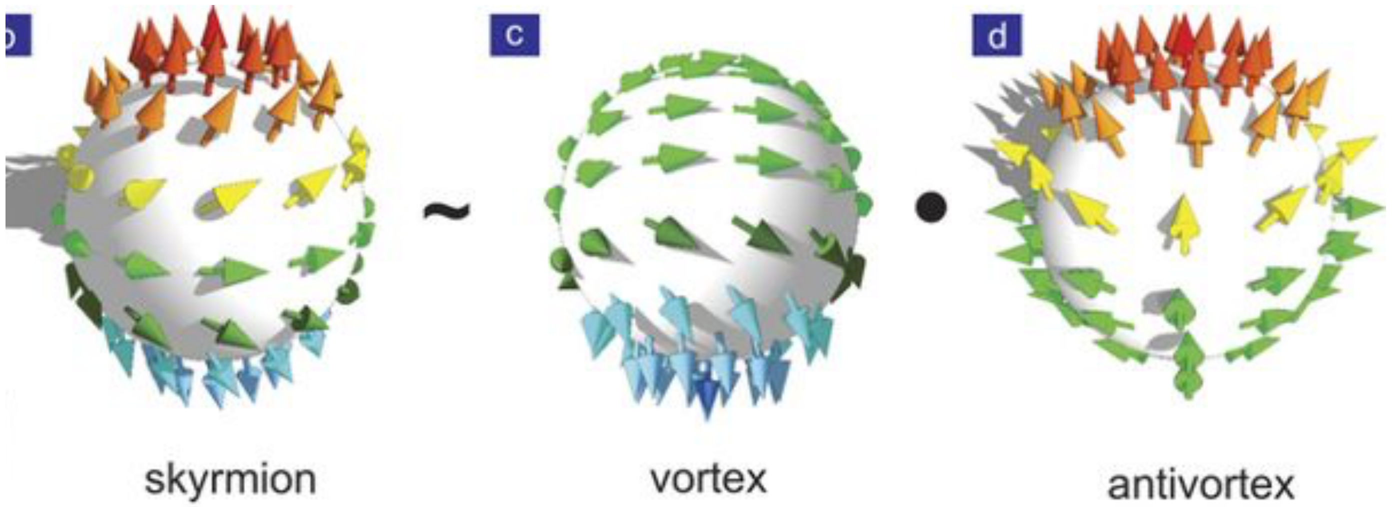

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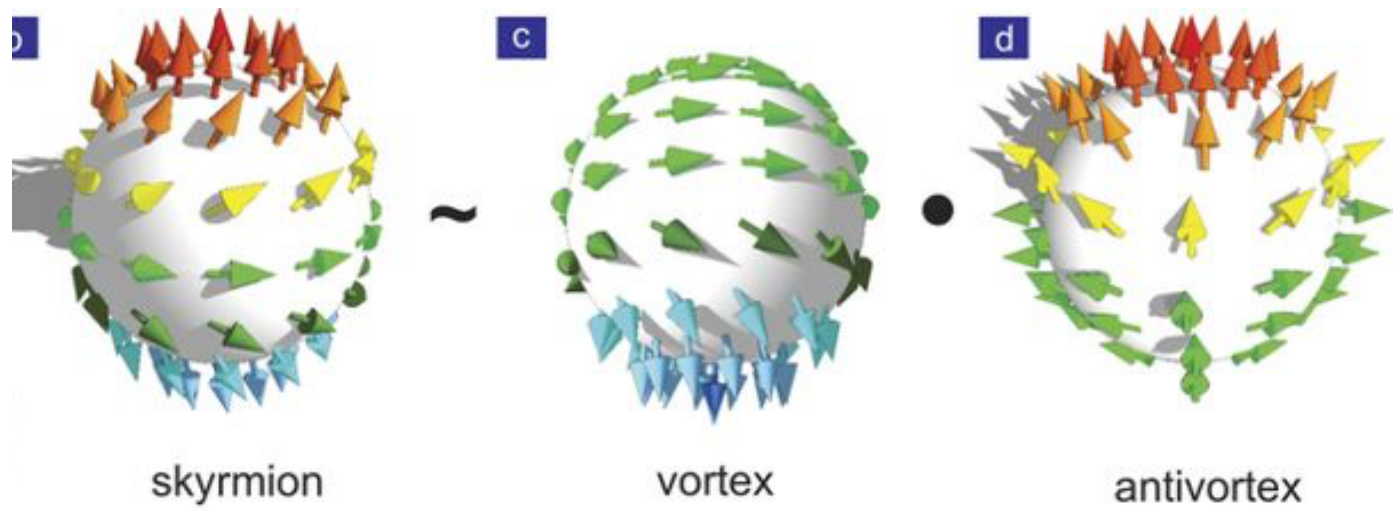

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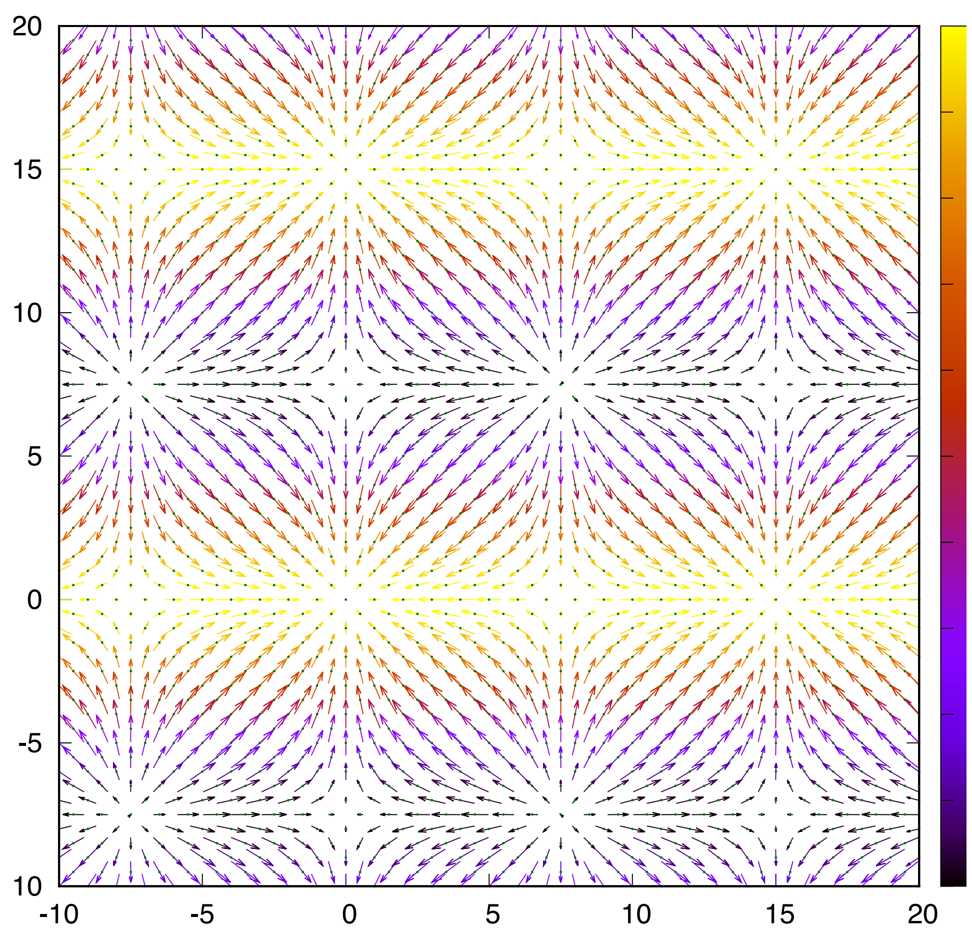
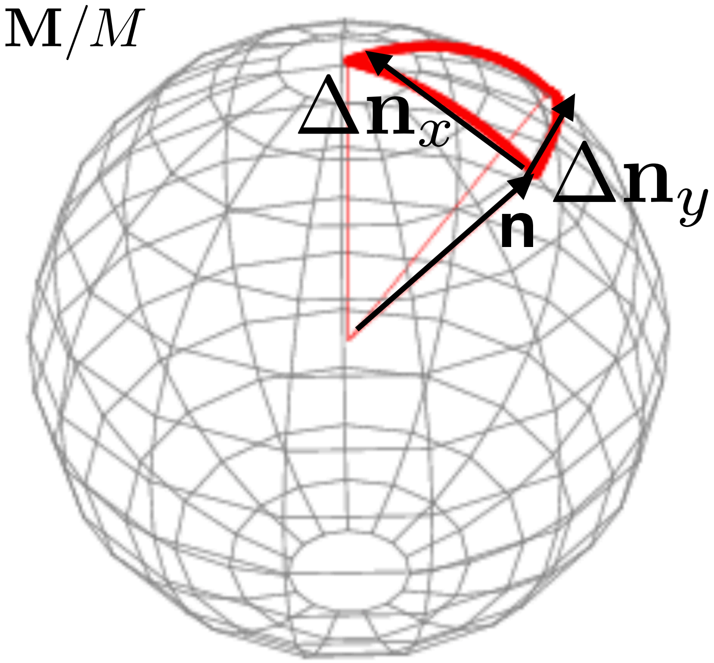
Artificial quasi-continuous magnetisation **M**(x,y)

(M_x,M_z) components in the xy plane, M_z-component by color



topological density/winding ~ solid angle

$$w(x,y) = \frac{1}{4\pi}(\mathbf{n}\cdot[\frac{\partial \mathbf{n}}{\partial x}\times\frac{\partial \mathbf{n}}{\partial y}]),\ \mathbf{n} = \mathbf{M}/M$$



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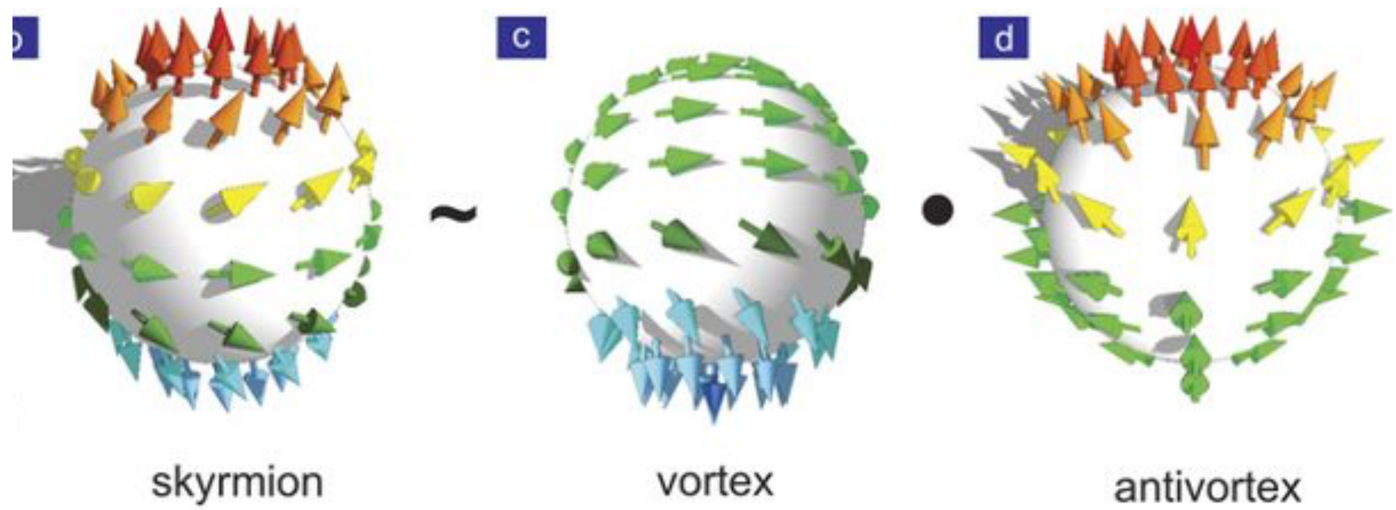

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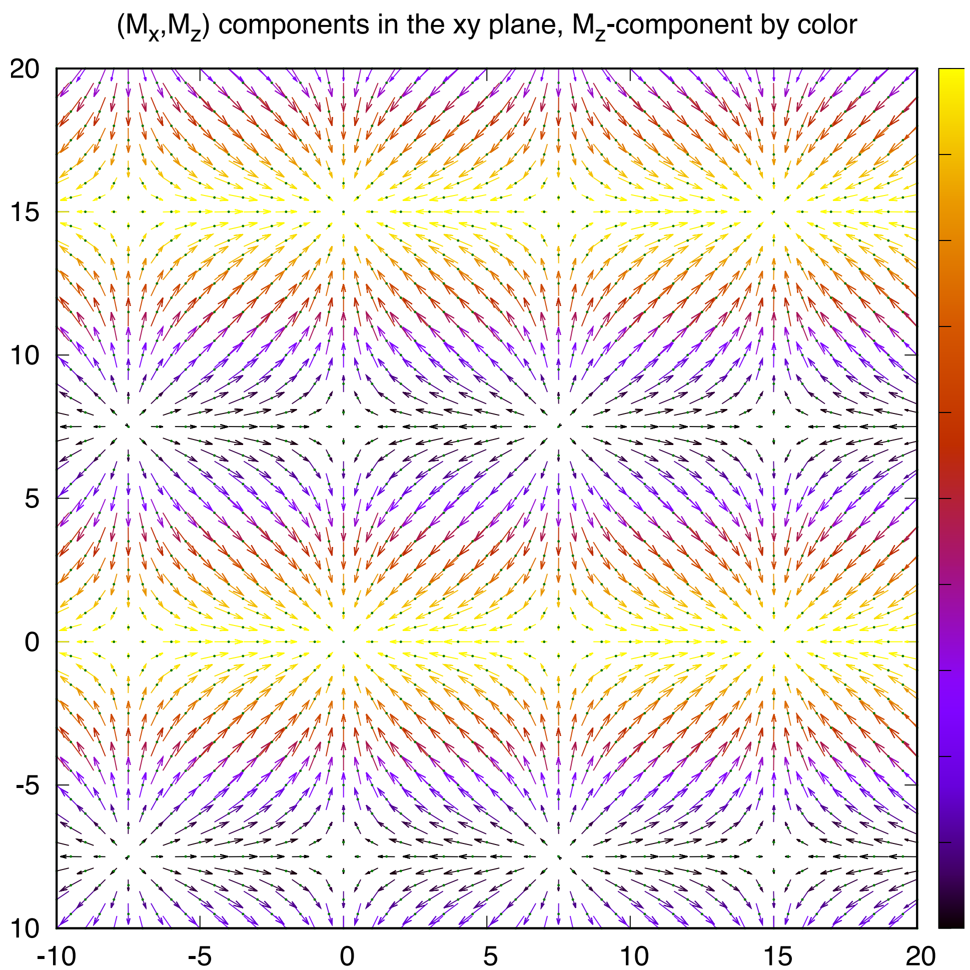

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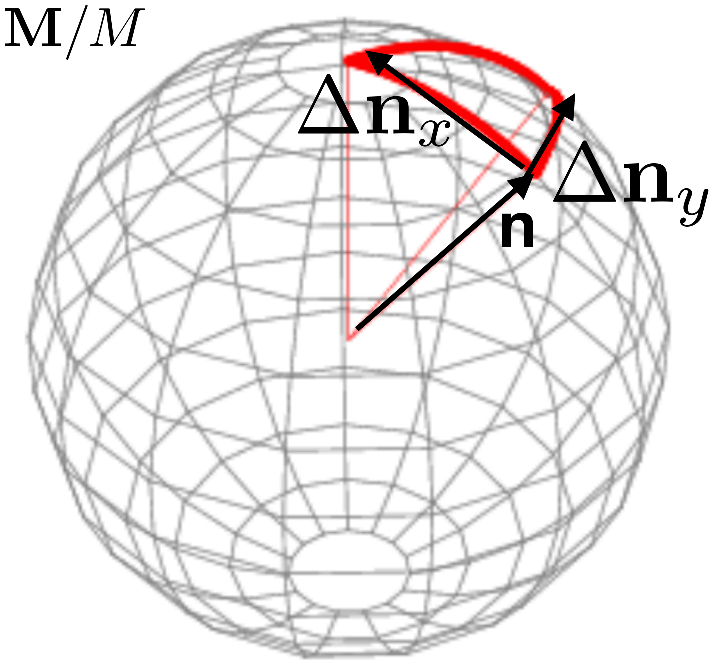
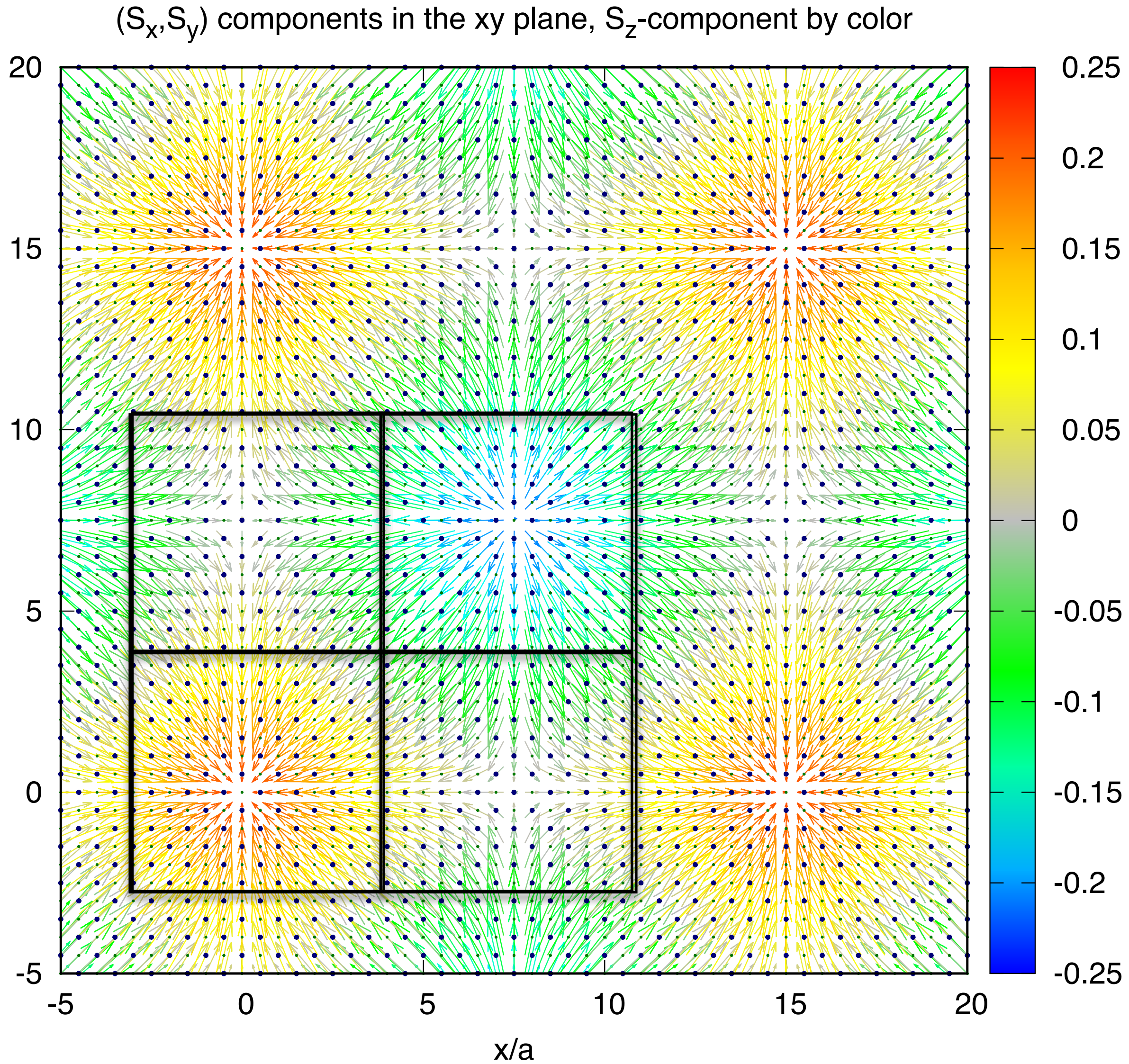


Topological number/charge

$$Q = \int\int w(x,y)dxdy$$

Topological skyrmion (Q=-1),
antiskyrmion (Q=+1) and
antimeron-meron (Q = ±1/2) for magnetization textures.

Continuous limit k->0 artificial full star magnetic structure



$$\mathbf{M}_{\mathrm{Ce1}} = m_1 \sin(x)\mathbf{e}_{\boldsymbol{x}} + m_2 \sin(y)\mathbf{e}_{\boldsymbol{y}} + \left(m_3 \cos(x) + m_4 \cos(y)\right)\mathbf{e}_{\boldsymbol{z}}$$

m1=m2=2 and m3=0.1, m4=0.11

$$\mathbf{M}_{\mathrm{Ce2}} = m_2 \sin(x)\mathbf{e}_{\boldsymbol{x}} + m_1 \sin(y)\mathbf{e}_{\boldsymbol{y}} + \left(m_4 \cos(x) + m_3 \cos(y)\right)\mathbf{e}_{\boldsymbol{z}}$$

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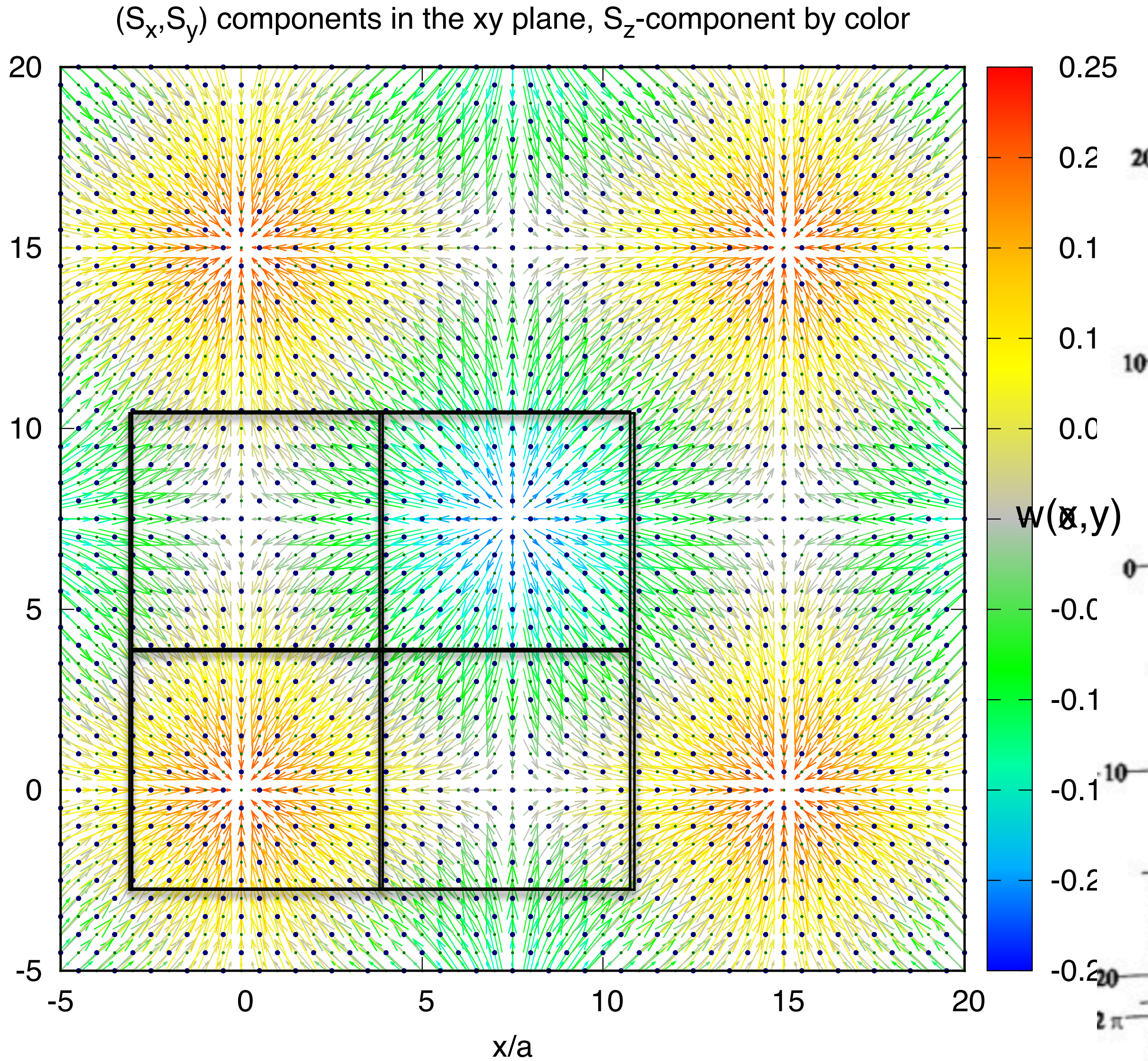

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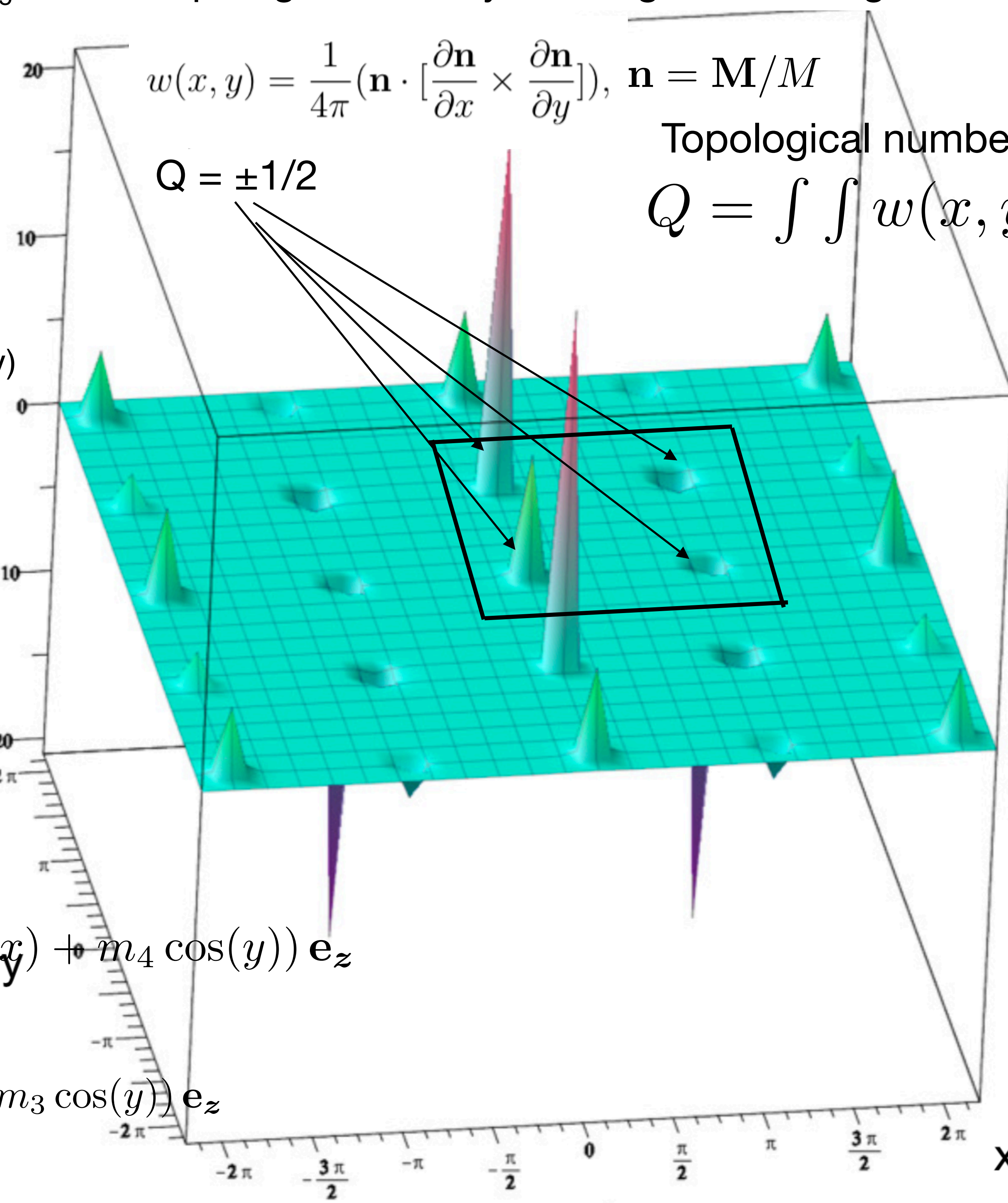

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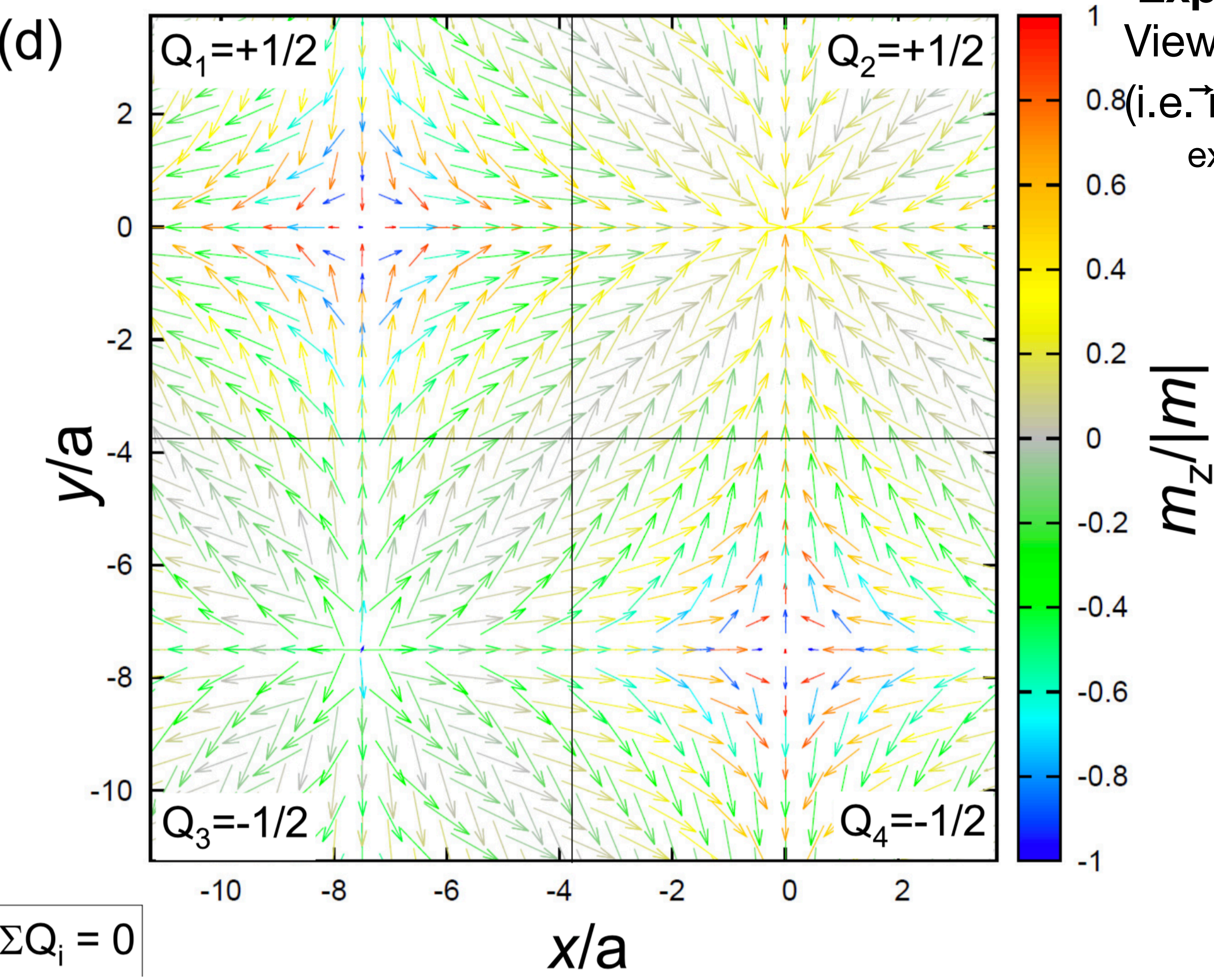
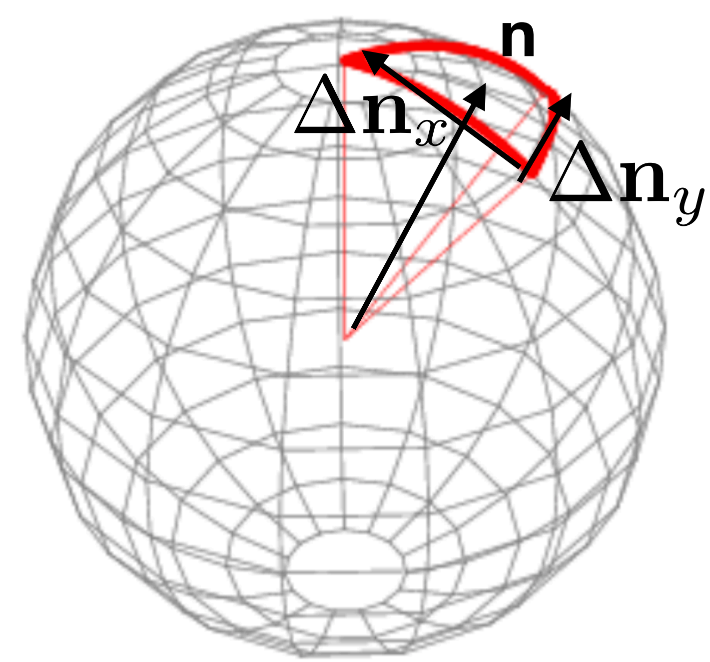
Our real case: magnetic meron in CeSlGe

$\mathbf{n} = \mathbf{M}/M$



$\Sigma Q_i = 0$

Experimentally observed multi-k magnetic structure.
View along the z-(c-)axis of the normalized
(i.e. $\vec{n} = \vec{M}/|\vec{M}|$, where $\vec{M}$ is the local Ce moment)
experiment: (m1,m2,m3,m4) = (0.44(1), 1.02(1), −0.21(5), 0.29(7)) μB.



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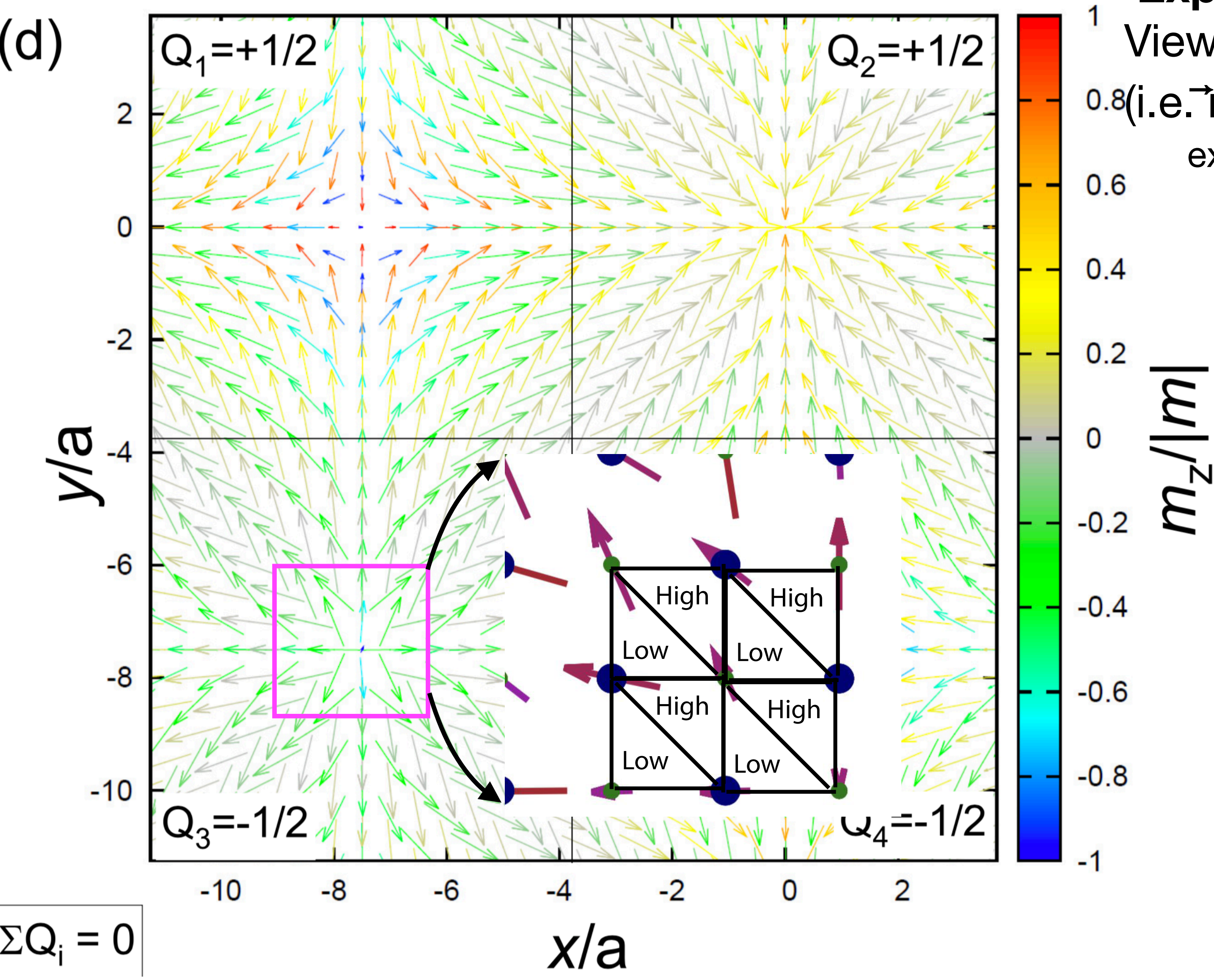

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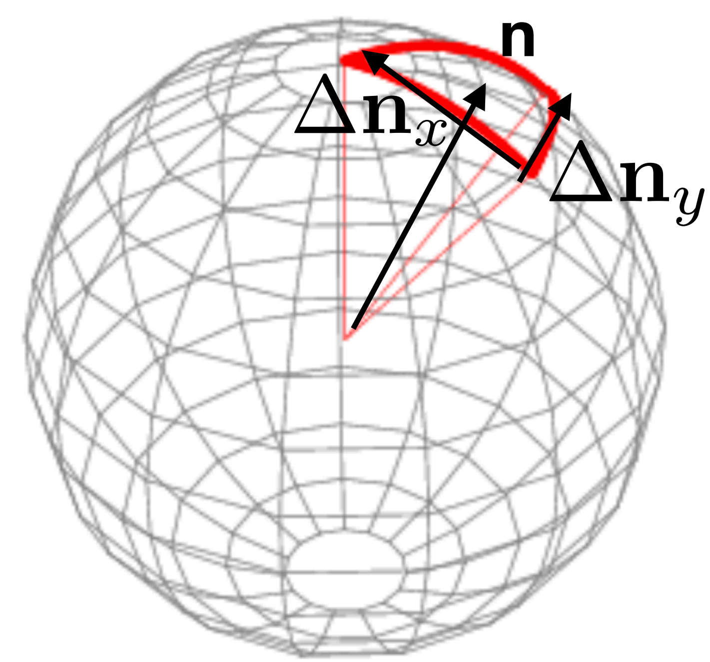


$\Sigma Q_i = 0$

topological density/winding ~ solid angle → $\Delta w(x,y) = \frac{1}{4\pi}\left(\mathbf{n}\cdot[\Delta\mathbf{n}_x \times \Delta\mathbf{n}_y]\right)^{☆}$

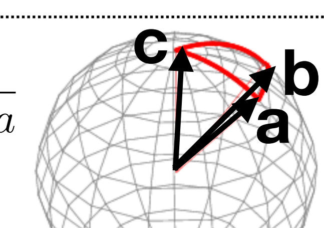
solid angle per square placket

$$w(x,y) = \frac{1}{4\pi}(\mathbf{n}\cdot[\frac{\partial\mathbf{n}}{\partial x}\times\frac{\partial\mathbf{n}}{\partial y}]),\ \mathbf{n} = \mathbf{M}/M$$

Topological number/charge → $Q = \sum_{x,y}\Delta w(x,y)$

$$Q = \int\int w(x,y)dxdy$$

☆ $\tan\left(\frac{1}{2}\Omega\right) = \frac{\mathbf{a}\cdot(\mathbf{b}\times\mathbf{c})}{abc + (\mathbf{a}\cdot\mathbf{b})\,c + (\mathbf{a}\cdot\mathbf{c})\,b + (\mathbf{b}\cdot\mathbf{c})\,a}$



Topological density and charge. H=0

experiment: (m1,m2,m3,m4) = (0.44(1), 1.02(1), −0.21(5), 0.29(7)) μB.

$$\Delta w(x,y) = \frac{1}{4\pi}\left(\mathbf{n}\cdot[\Delta\mathbf{n}_x \times \Delta\mathbf{n}_y]\right)$$

solid angle per square placket

$$Q = \sum_{x,y} \Delta w(x,y)$$

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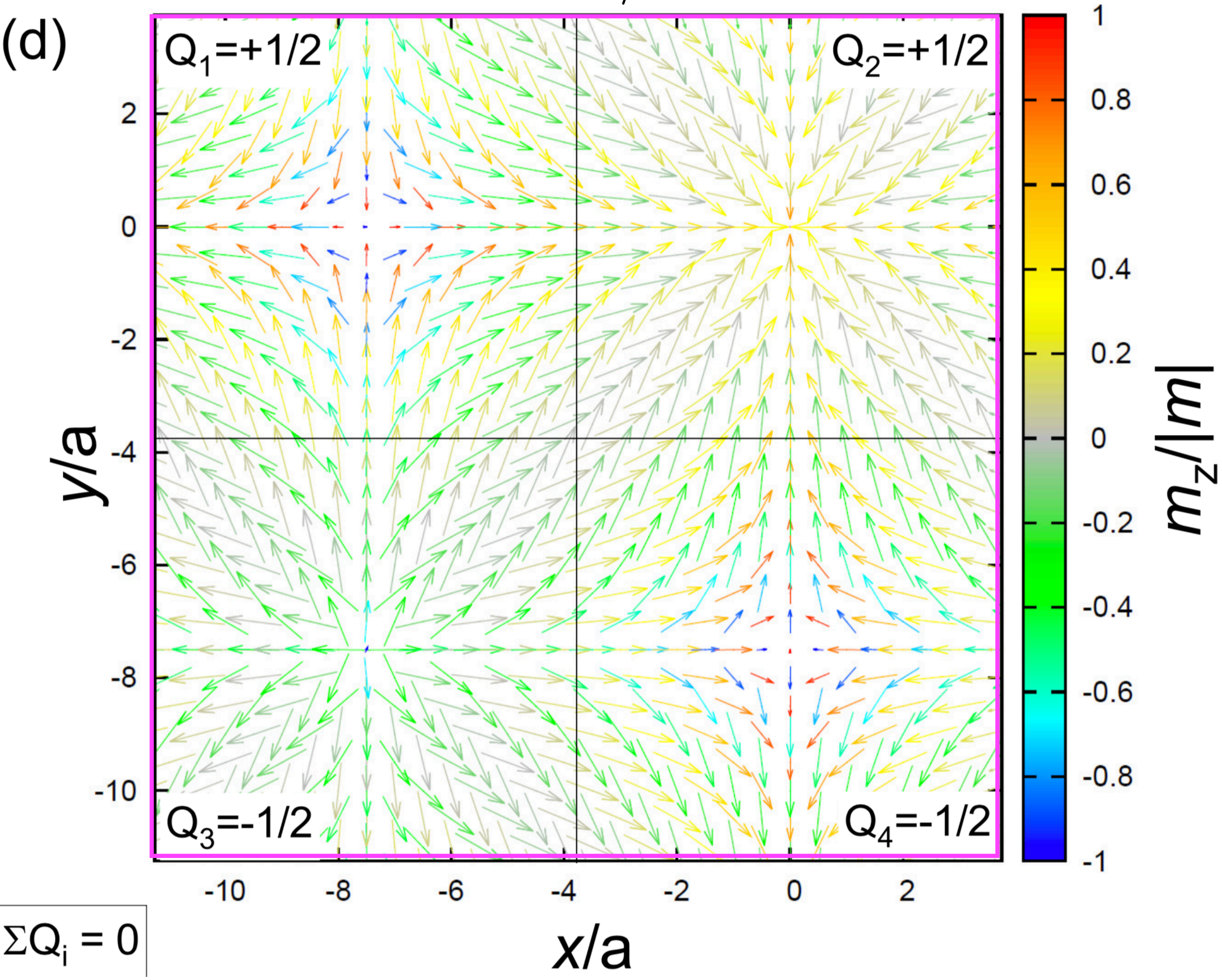

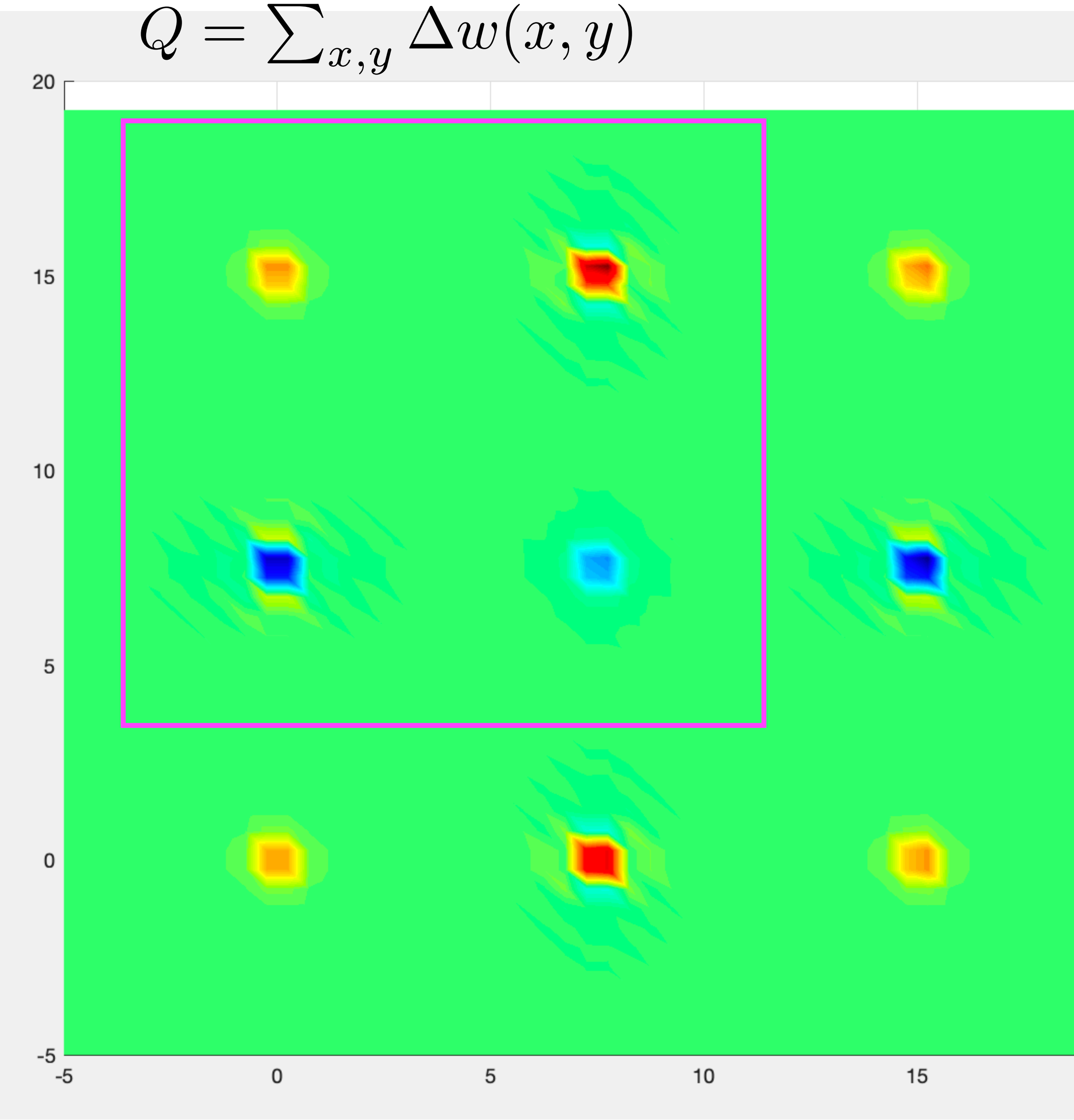

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Simulation of external field. FM component along z-axis

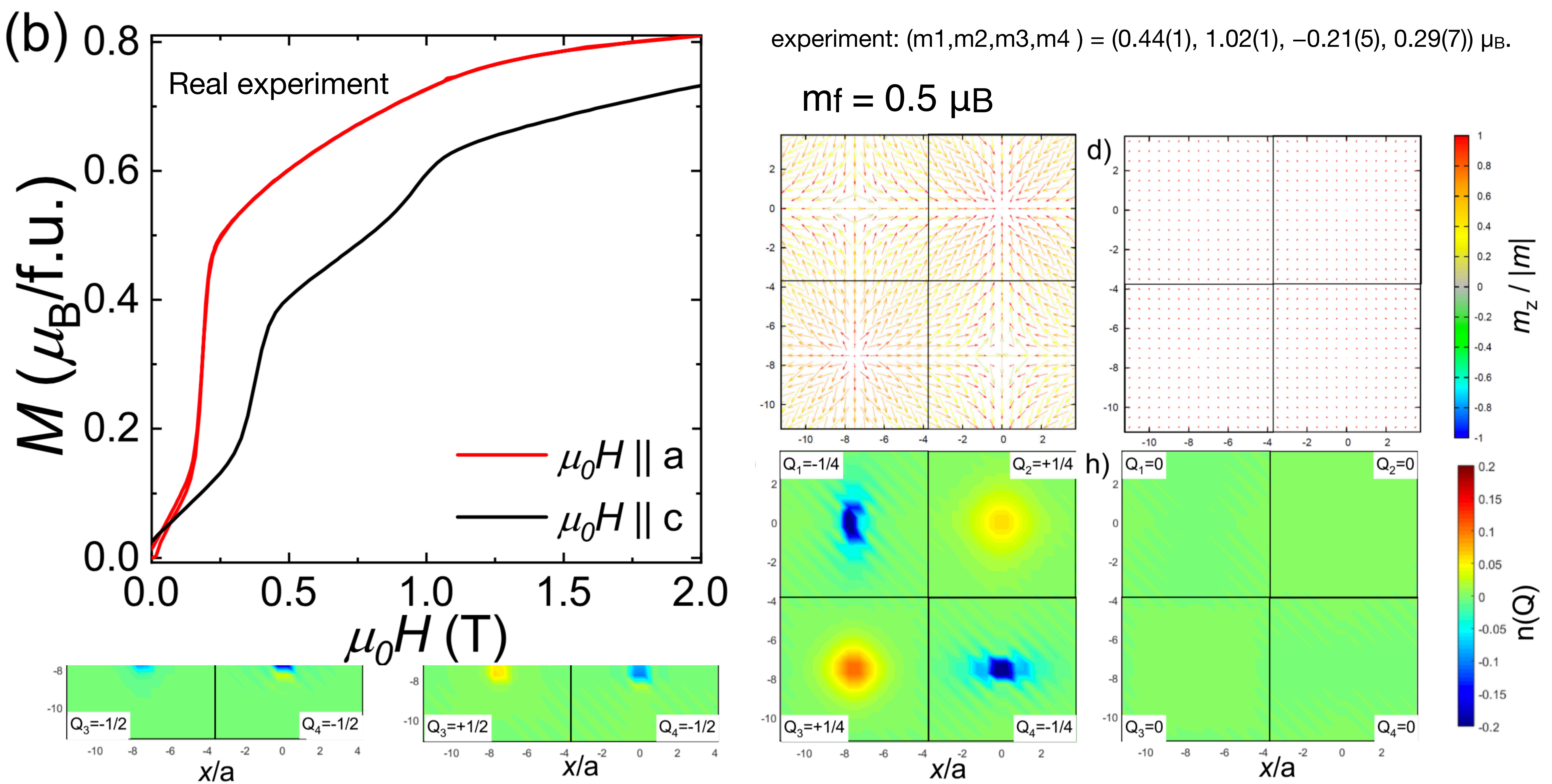

Figure 10. Comparison between the normalized moment $\vec{M}/|\vec{M}|$ and winding density for increasing canting fields along th z-direction out of the page. The steps of fields are m_f= a,e) 0 μ_B b,f) 0.2 μ_B c,g) 0.5 μ_B and d,h) 10 μ_B. a-d) The first row o mages shows the view along the z-(c-)axis of the magnetic order. The cases for m_f = 0 μ_B (a) and m_f = 0.2 μ_B (b) are th same ones shown in Figure 1(d) and Figure 5 of the main text. The x- and y-axes are in units of in-plane lattice parameter

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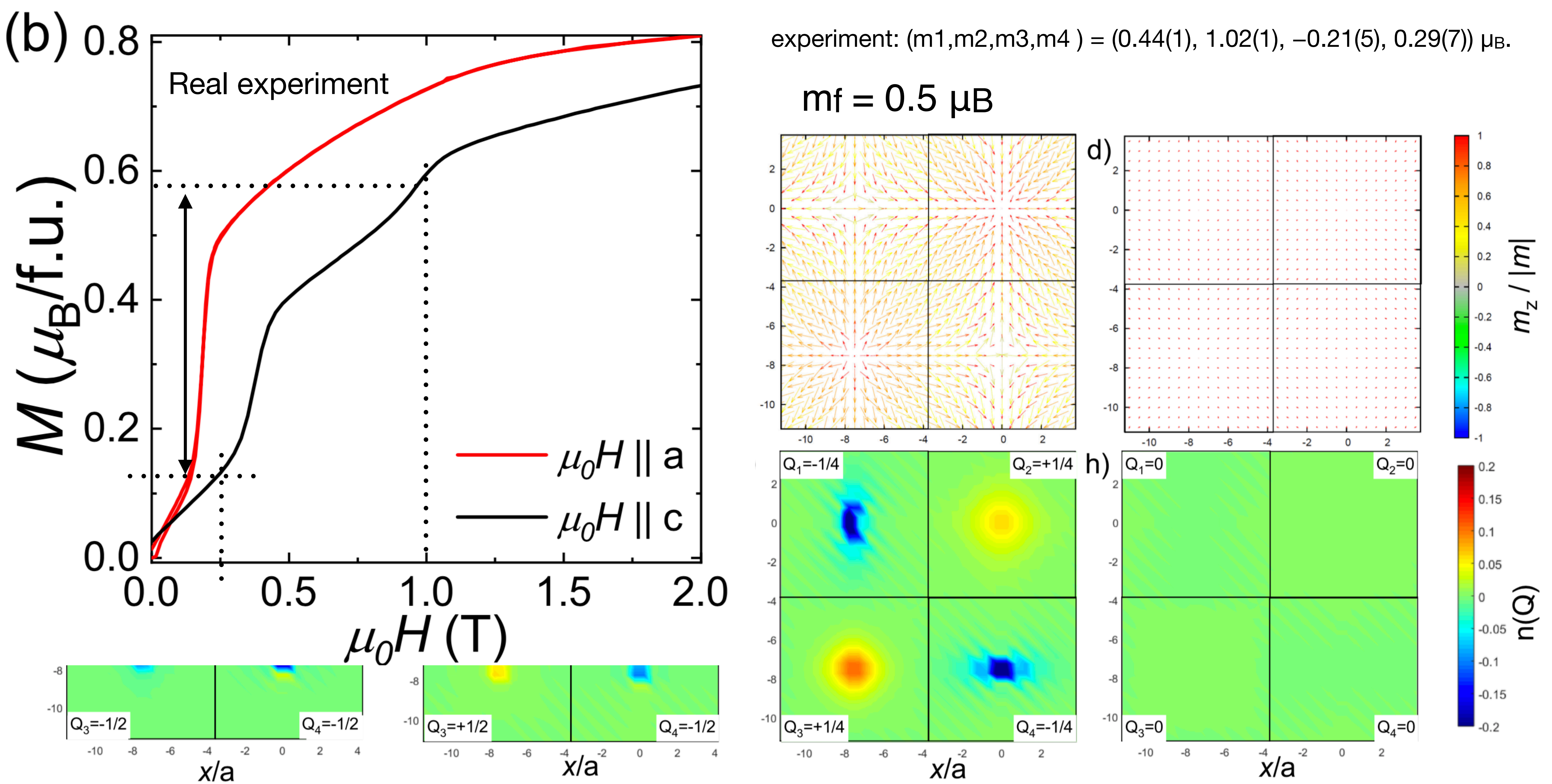

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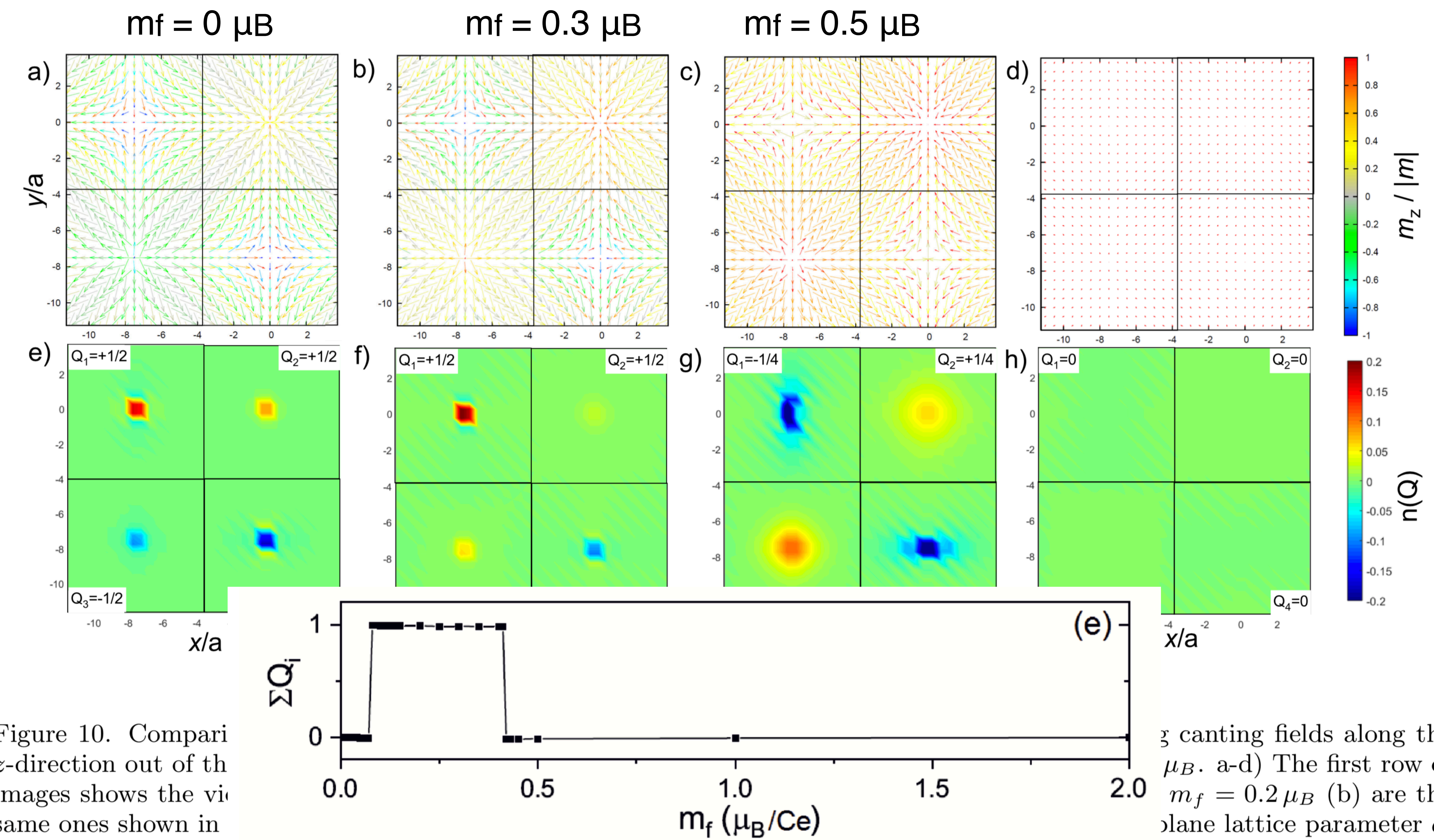
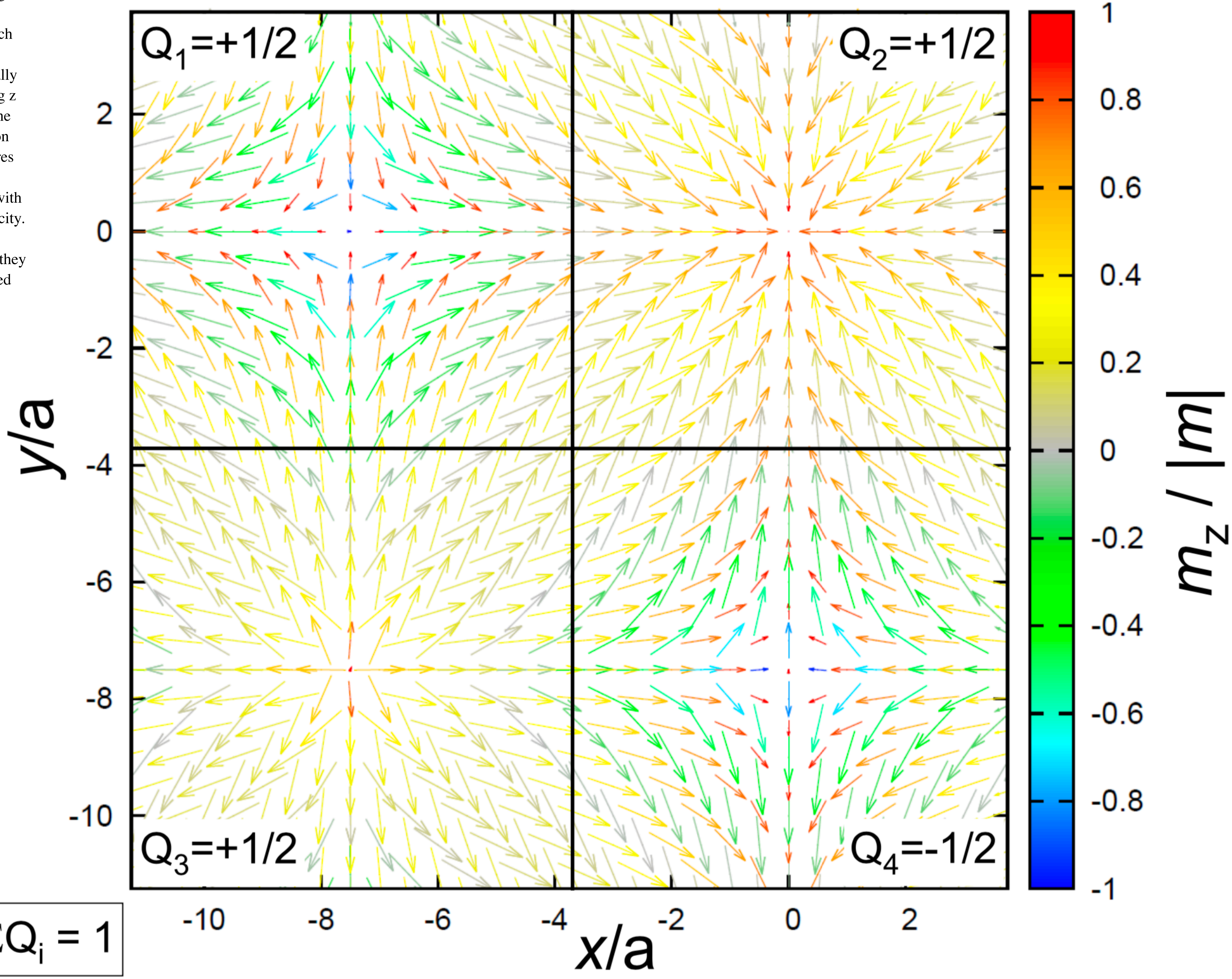


Figure 10. Compari… canting fields along th… z-direction out of th… μ_B. a-d) The first row … images shows the vi… $m_f = 0.2\,\mu_B$ (b) are th… same ones shown in … plane lattice parameter …

“intermediate field” FM component along z -> total charge Q=1

At low mf =0.08 μB /Ce, we find ∑ Qi changes sharply from 0 to +1, with the resulting topological order. Unlike skyrmions for which the spin texture wraps a sphere and Q=|1|, merons and antimerons are described generally by a magnetization aligned up or down along z in a core region, that gradually aligns in-plane towards the edge. Following the classification scheme given in Refs. 23, 44, the spin textures in the top-right (Q2) and bottom-left (Q3) quadrants shown in Figure 5 are consistent with being core-up antimerons with opposite helicity. The remaining quadrants ((Q1 and (Q4)) in Figure 5 are different to (anti)merons, since they contain moments both aligned and antialigned with z.



$\Sigma Q_i = 1$

Summary

- We report the discovery of topological magnetic order in the polar tetragonal magnetic Weyl semimetal candidate CeAlGe.

- CeAlGe is a host of incommensurate magnetic structure below T_N, [3D+2 group *I4_1md1'(a,0,0)000s(0,a,0)0s0s*] hosting a lattice of magnetic particle-like objects called antimerons Q=1/2 with half-integer topological numbers.

- The topological properties - two antimerons with total Q=1 of the phase stable at intermediate magnetic fields parallel to the c-axis is confirmed by the observation of a topological Hall effect (THE).

P. Puphal, et al, submitted (2019)

END