The quest for the propagation vector

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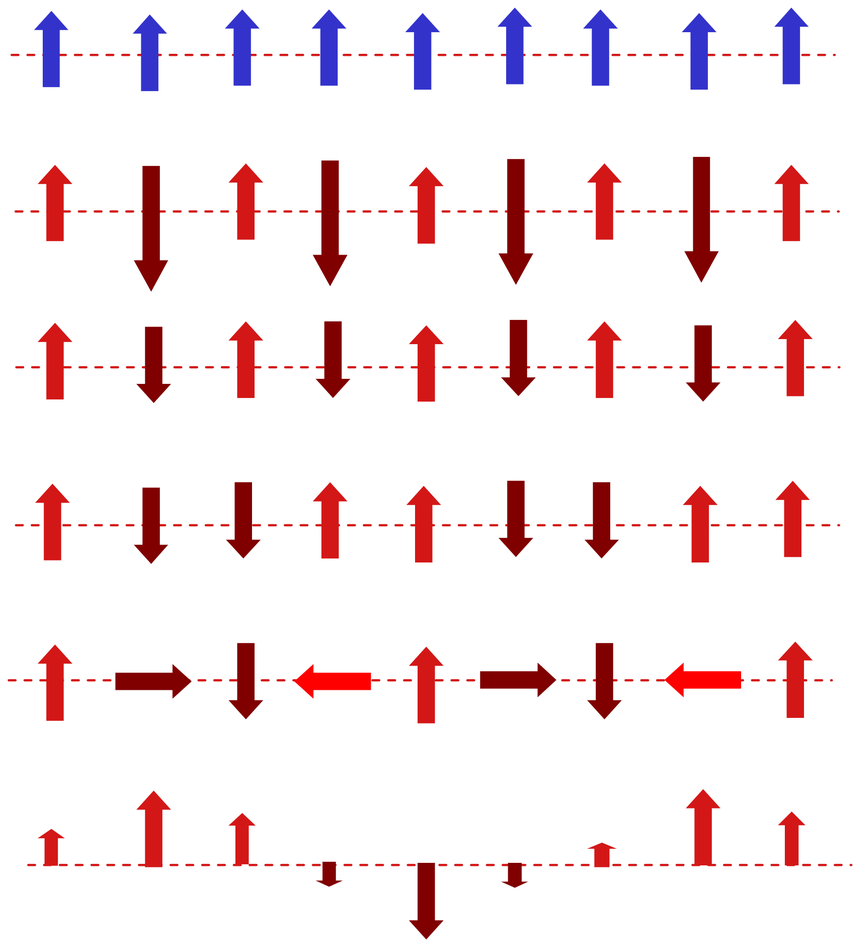
Magnetic structure

Where the propagation vector comes in

- Exchange coupling -> ordering of moments
- Magnetic anisotropy -> moment orientation
- What characterises a magnetic structure ?
  - Propagation vector(s)
  - Magnitude of moments
  - Direction of moments
- Interactions with neutrons
  - Nuclear interaction between neutrons and the nuclei
  - Magnetic interaction between neutron’s magnetic moment with (spin and orbital) magnetic moments of the unpaired electrons in condensed matter

Types of magnetic structure models in 1D



- Ferromagnet
- Collinear &
- Antiferromagnet
- Antiferromagnet
- Non-collinear antiferromagnet
- Incommensurate antiferromagnet

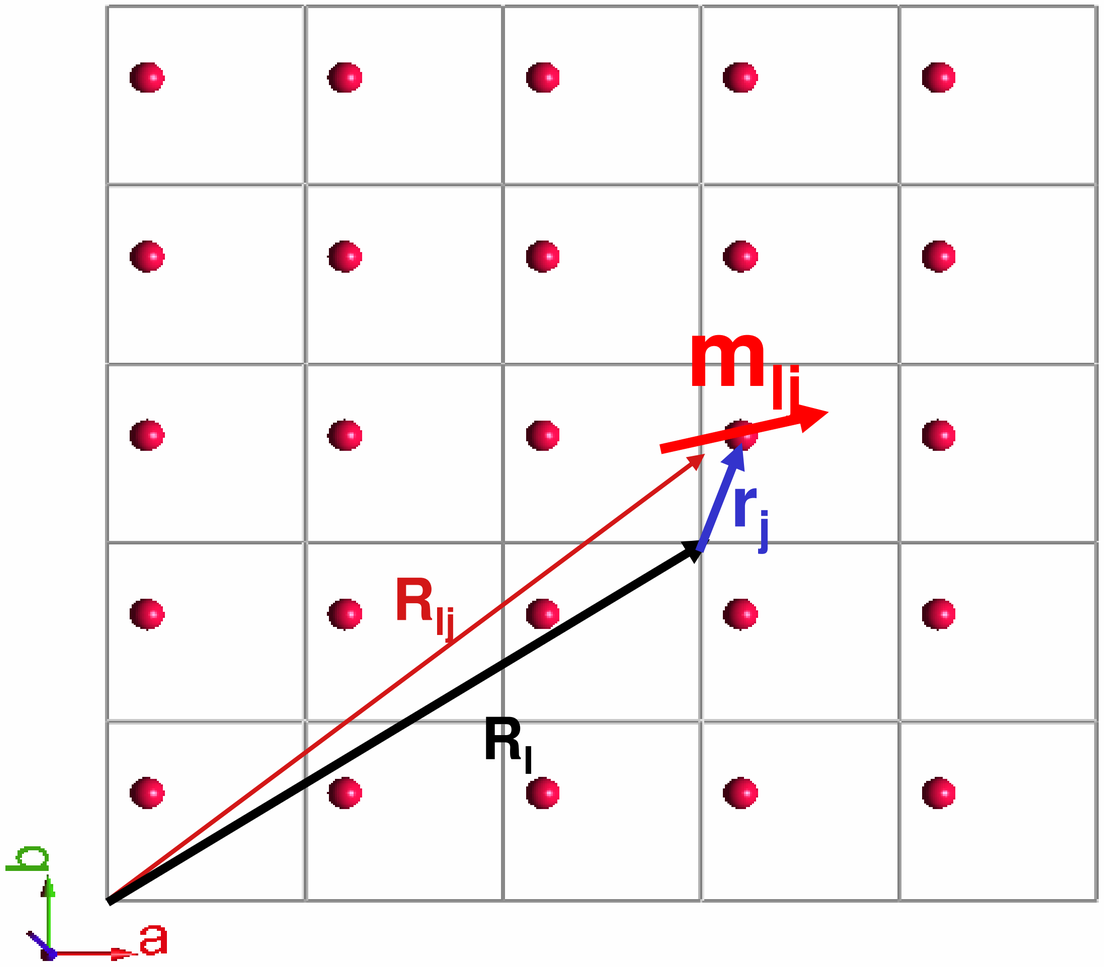
Propagation vector

The concept

- Commensurate magnetic structures can be described by a periodic repetition of a magnetic unit cell
  - Just as for (nuclear) crystal structures by a repetition of a nuclear unit cell
- One way of describing the magnetic unit cell:
  - Based on the nuclear unit cell
  - **Propagation vector** (**k**)
    - Relation between moment orientations of **equivalent** magnetic atoms in **different** nuclear unit cells

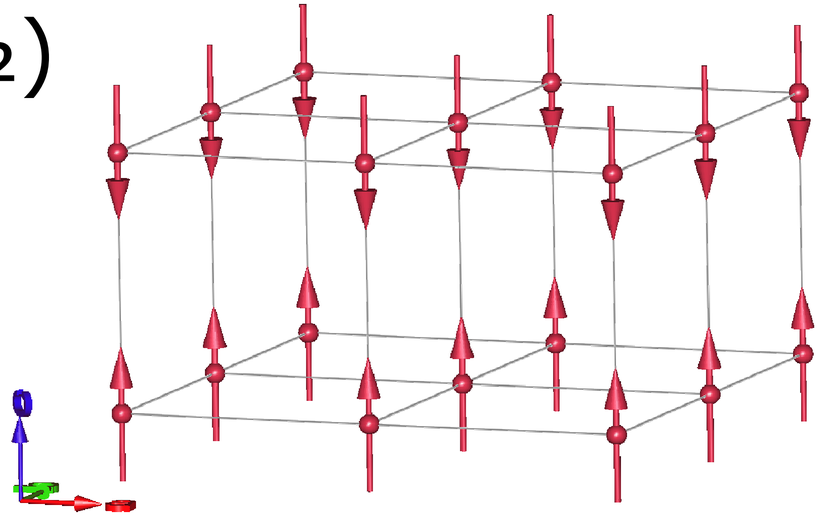
Propagation vector

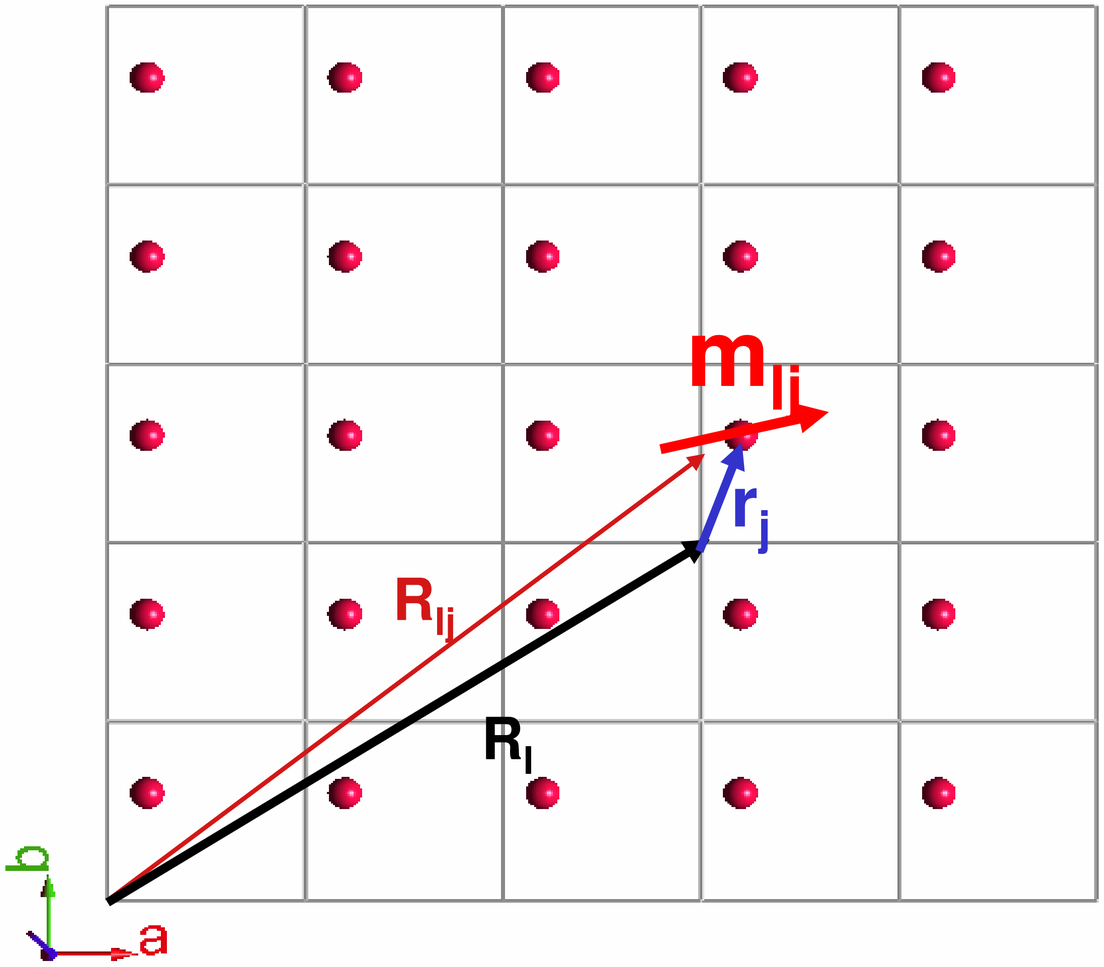
- Atom j in unit cell l
  - $\mathbf{R}_{lj}=\mathbf{R}_l+\mathbf{r}_j$
- Fourier series:
  - $\mathbf{m}_{lj} = \sum_{\mathbf{k}}[\mathbf{S}_{\mathbf{k}j} \exp(-2\pi i\mathbf{k}\mathbf{R}_l)]$
  - $\mathbf{S}_{\mathbf{k}j}$ Fourier components
    - Linear combination of basis vectors of irreducible representations
    - $\mathbf{S}^*_{\mathbf{k}j} = \mathbf{S}_{-\mathbf{k}j}$ for $\mathbf{m}_{lj}$ to be real



Propagation vector

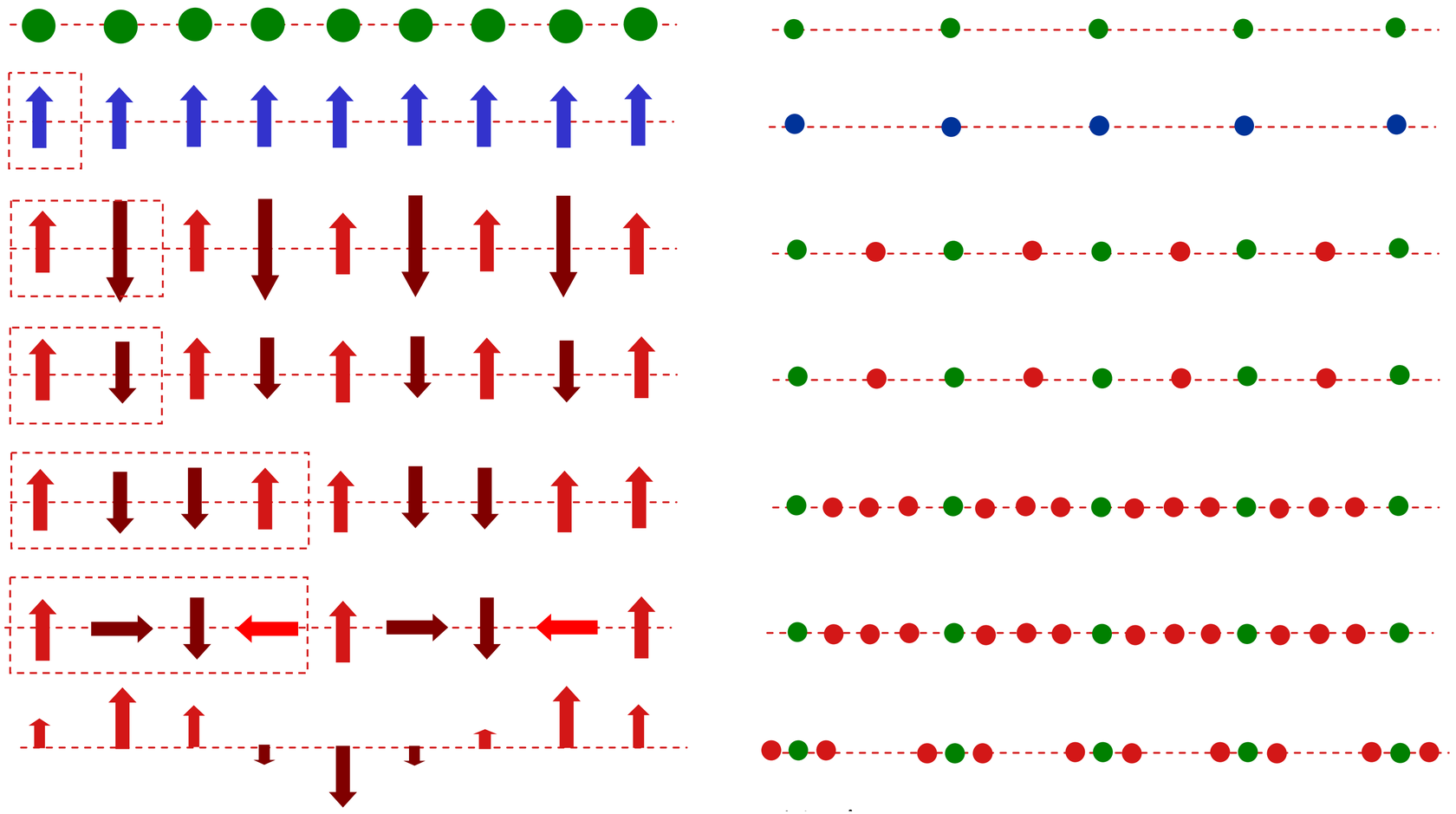
- Atom *j* in unit cell *l*
  - $\mathbf{R}_{lj}=\mathbf{R}_l+\mathbf{r}_j$
- For one propagation vector:
  - $\mathbf{m}_{lj} = \mathbf{S}_{\mathbf{k}j} \exp(-2\pi i\mathbf{k}\mathbf{R}_l)$
  - $\mathbf{k}=(00½)$





Diffraction of magnetic structure models in 1D

Back to the simple models…

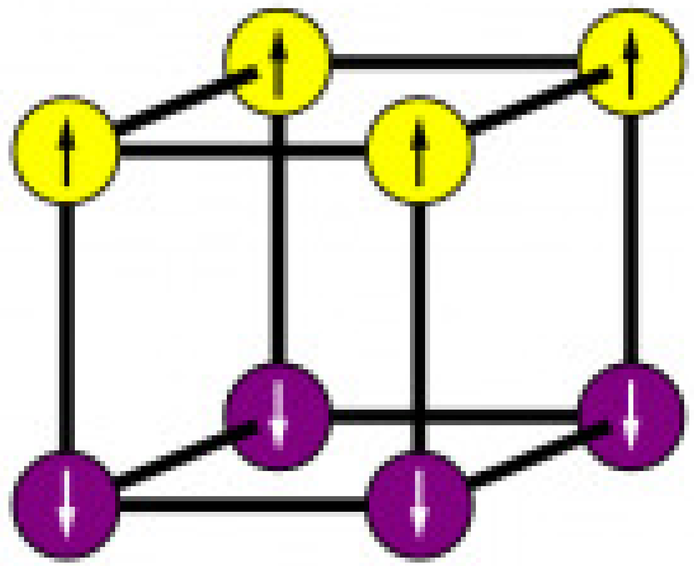


- Paramagnet
- Ferromagnet
- Collinear ferrimagnet
- Antiferromagnet
- Antiferromagnet
- Non-collinear AFM
- Incommensurate AFM

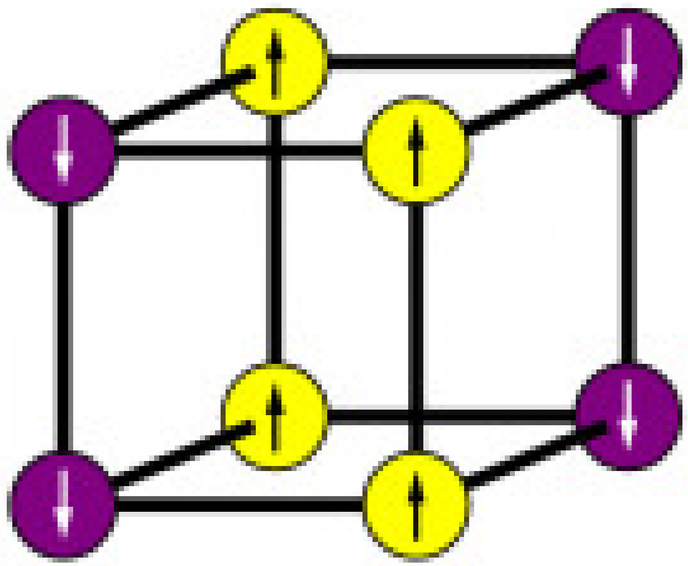
Propagation vector examples in 3D

Supposing a magnetic atom in the corner of the unit cell

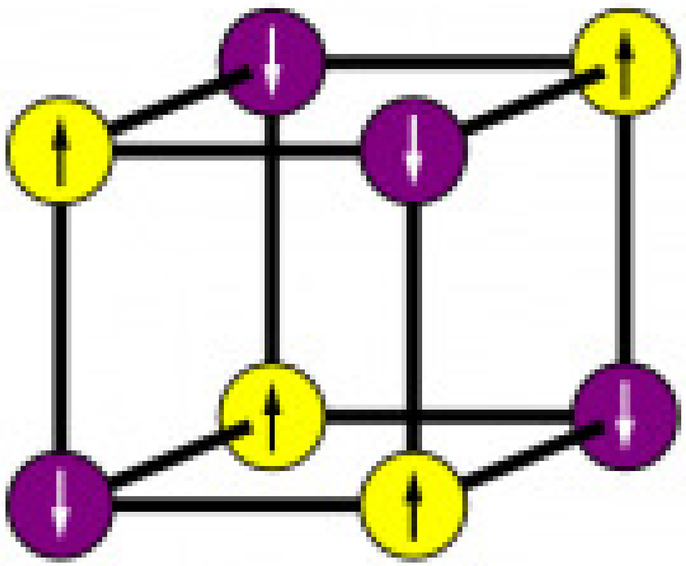
- **k**=(00½)



k=(½½0)



k=(½½½)



Calculate a magnetic structure

Let's se what magics the propagation vector can do…

- Propagation vectors $\mathbf{k}=(¼ ¼ ½)$, $-\mathbf{k}=(-¼ -¼ -½)$
- $\mathbf{S_k}=(0,0,\mu(1+i)/2)$ $\mathbf{S}^*_\mathbf{k}=\mathbf{S_{-k}}=(0,0,\mu(1-i)/2)$
- One atom only per unit cell
- $\mathbf{m}_l= \Sigma_\mathbf{k}[\mathbf{S}_{\mathbf{k}j} \exp(-2\pi i\mathbf{kR}_l)]$
  $=(0,0,\mu(1+i)/2)\exp(-2\pi i\mathbf{kR}_l)+(0,0,\mu(1-i)/2)\exp(-2\pi i\mathbf{kR}_l)$
- $\mathbf{S_k}=½(\mathbf{R_k}+i\mathbf{I_k})$ with $\mathbf{R_k}=\mathbf{I_k}=(00\mu)$
- $\mathbf{m}_l=\mathbf{R_k}\cos(2\pi\mathbf{kR}_l)+\mathbf{I_k}\sin(2\pi\mathbf{kR}_l)$

Calculate a magnetic structure

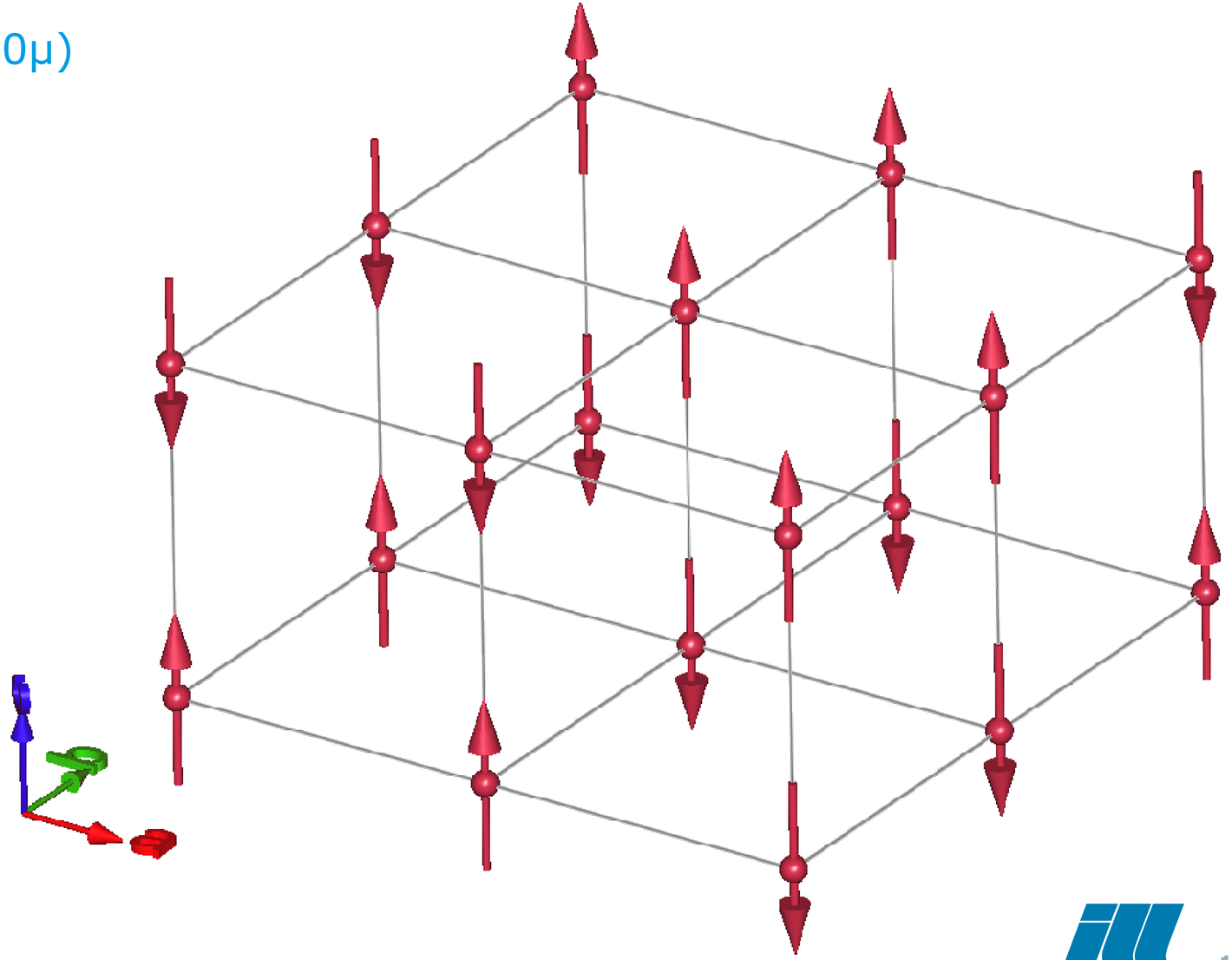
$\mathbf{k}=(¼¼½)$, $\mathbf{S_k}=½(\mathbf{R_k}+i\mathbf{I_k})$ with $\mathbf{R_k}=\mathbf{I_k}=(00\mu)$

- $\mathbf{m}_{000}=(00\mu)\cos(2\pi(¼¼½)(000))+(00\mu)\sin(2\pi(¼¼½)(000))$
  $=(00\mu)$
- $\mathbf{m}_{100}=(00\mu)\cos(2\pi(¼¼½)(100))+(00\mu)\sin(2\pi(¼¼½)(100))$
  $=(00\mu)(\cos(\pi/2)+\sin(\pi/2))=(00\mu)$
- $\mathbf{m}_{200}=(00\mu)\cos(2\pi(¼¼½)(200))+(00\mu)\sin(2\pi(¼¼½)(200))$
  $=(00\mu)(\cos(\pi)+\sin(\pi))=(00\text{-}\mu)$
- $\mathbf{m}_{300}=(00\mu)\cos(2\pi(¼¼½)(300))+(00\mu)\sin(2\pi(¼¼½)(300))$
  $=(00\mu)(\cos(3\pi/2)+\sin(3\pi/2))=(00\text{-}\mu)$
- $\mathbf{m}_{001}=(00\mu)\cos(2\pi(¼¼½)(001))+(00\mu)\sin(2\pi(¼¼½)(001))$
  $=(00\mu)(\cos(\pi)+\sin(\pi))=(00\text{-}\mu)$
- $\mathbf{m}_{002}=(00\mu)\cos(2\pi(¼¼½)(002))+(00\mu)\sin(2\pi(¼¼½)(002))$
  $=(00\mu)(\cos(2\pi)+\sin(2\pi))=(00\mu)$

Calculate a magnetic structure

$\mathbf{k}=(¼\,¼\,½)$, $\mathbf{S_k}=½(\mathbf{R_k}+i\mathbf{I_k})$ with $\mathbf{R_k}=\mathbf{I_k}=(00\mu)$

- $\mathbf{m}_{000}=(00\mu)$
- $\mathbf{m}_{100}=(00\mu)$
- $\mathbf{m}_{200}=(00\text{-}\mu)$
- $\mathbf{m}_{300}=(00\text{-}\mu)$
- $\mathbf{m}_{001}=(00\text{-}\mu)$
- $\mathbf{m}_{002}=(00\mu)$



How to find the propagation vector?

Let's be pragmatic, go through an example

- Nuclear structure known (at least unit cell)
- 2 diffraction patterns, (just) above and (well) below transition temperature
- Rietveld (or Le Bail) fit of the low temperature pattern with nuclear structure model from high temperature
- Indexing exercise on remaining peaks' positions
  - In contrast to indexing of a completely unknown nuclear structure pattern there's a relation to the known unit cell already, we need just to find a simple vector (well… sometimes more than one…) which means less degrees of freedom
  - At least for commensurable magnetic ordering, one does not search in a limitless space of possibilities, but the vector components are likely simple rational numbers n/m with $n \in \{0,1,2,3,\ldots\}$ and $m \in \{2,3,\ldots\}$

$Sr_4NaRu_3O_{12}$

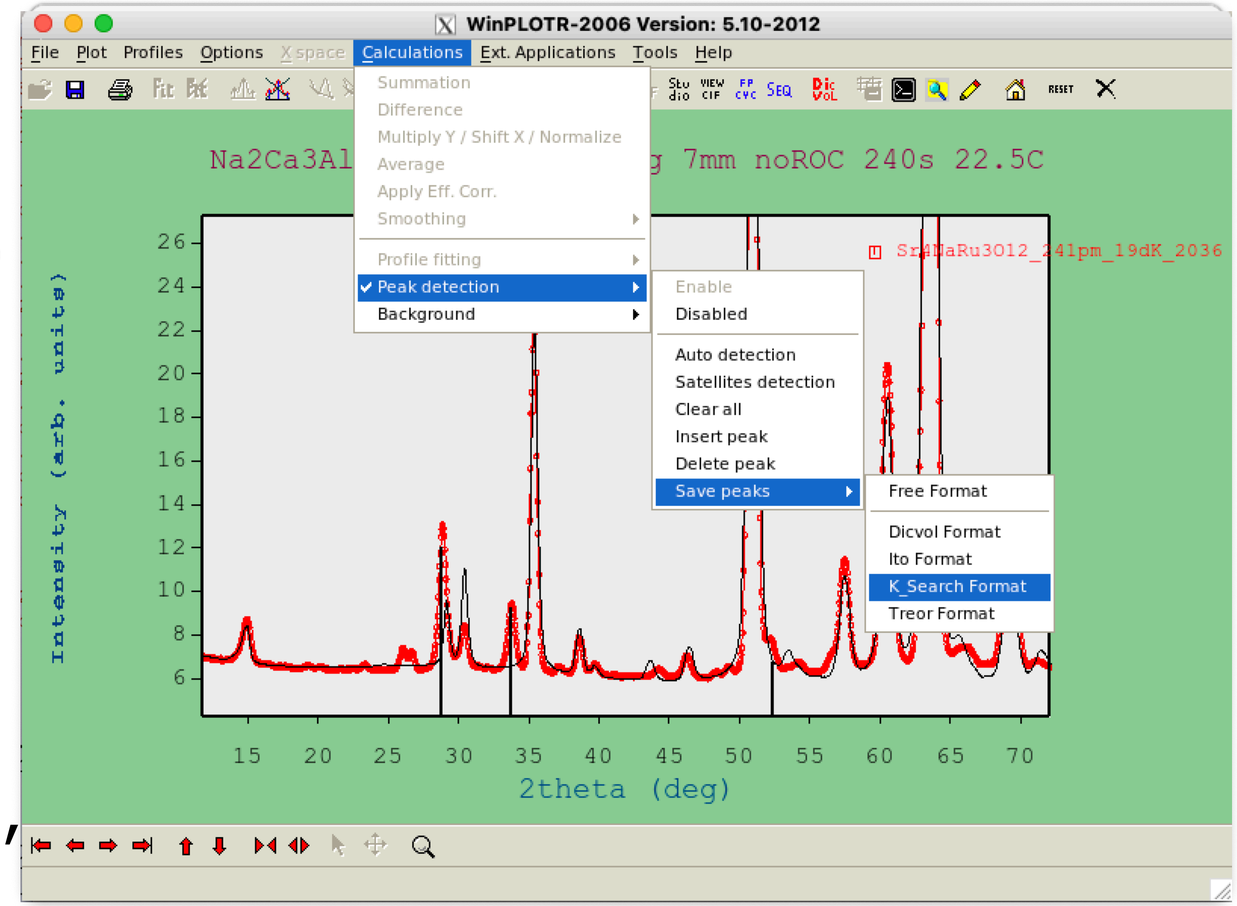
Not yet published - please do not copy 😆 !

- Structure from single-xtl XRD and HiRes-NPD
- Neutron powder diffraction data from D20
  - relatively long wavelength (2.41Å),
  - low take-off angle (42º)
  - to have high intensity and resolution where it counts for magnetism
- Structure model refined (only overall-Biso, zero-shift and unit cell) with paramagnetic phase at 2.41Å (275K)
- Same refinement with low-T data (1.9K)
- Look at it with WinPlotR-2006…

$Sr_4NaRu_3O_{12}$

WinPlotR-2006 and k_search

- Search satellite peaks
  - A very small number will do!
  - Check carefully if they really belong to the magnetic phase
  - Consider impurity peaks
- Be very careful with zero-shift, tolerances etc.
  - Add zero-shift to obtained peak positions and reduce the tolerance
- Consider vector components >1, e.g., for rhombohedral system



k_search

It may be damned difficult to find the right solution!

```
! Input file for K_SEARCH
! created by WinPLOTR-2006:30/ 1/2023 at 13:49:29
! Data file : processed/Sr4NaRu3O12_241pm_19dK_203666.prf
TITLE Na2Ca3Al2F14 Ge115 118deg 7mm noROC 240s 22.5C
SPGR     R -3
CELL      11.16420  11.16420  27.54107   90.0000   90.0000  120.0000
SHORT-OUTPUT
K_COMMENSURATE
CWTOL        2.40729    0.300
K_RANGE    0.0  0.5    0.0  0.5    0.0  0.5
K_DIV         100         100         100
! peak_position  peak_intensity  peak_background
2THETA_LIST      3
       28.85954       11.86837        6.66628
       33.82216        9.24502        5.72896
       52.36275        6.74802        5.82946
```

⇒ List of the best incommensurate 10 solutions for 3 satellites

Kx	Ky	Kz	R-factor
0.500000	0.250000	0.250000	0.110600
0.333330	0.500000	0.333330	0.242264
0.000000	0.125000	0.000000	0.302578
0.125000	0.000000	0.000000	0.302582
0.125000	0.250000	0.125000	0.315384
0.000000	0.500000	0.500000	0.327471
0.500000	0.500000	0.500000	0.327478
0.500000	0.333330	0.000000	0.406842
0.333330	0.500000	0.000000	0.406850
0.000000	0.500000	0.250000	0.458283

```
!  Zero    Code    SyCos    Code   SySin    Code  Lambda     Code MORE ->Patt# 1
  0.03655  161.0  0.00000    0.0  0.00000    0.0 2.407293    0.00   0
```

```
! Input file for K_SEARCH
! created by WinPLOTR-2006:30/ 1/2023 at 11: 7:47
! Data file : processed/Sr4NaRu3O12_241pm_19dK_203666.prf
TITLE Na2Ca3Al2F14 Ge115 118deg 7mm noROC 240s 22.5C
SPGR     R -3
CELL      11.16420  11.16420  27.54107   90.0000   90.0000
120.0000
SHORT-OUTPUT
K_COMMENSURATE
CWTOL        2.40729    0.300
K_RANGE    0.0  0.5    0.0  0.5    0.0  2.5
K_DIV         100         100         100
! peak_position  peak_intensity  peak_background
2THETA_LIST      2
       28.896       11.86836        6.35789
       33.8587       9.24502        6.51561
       52.3993       6.74802        5.82946
```

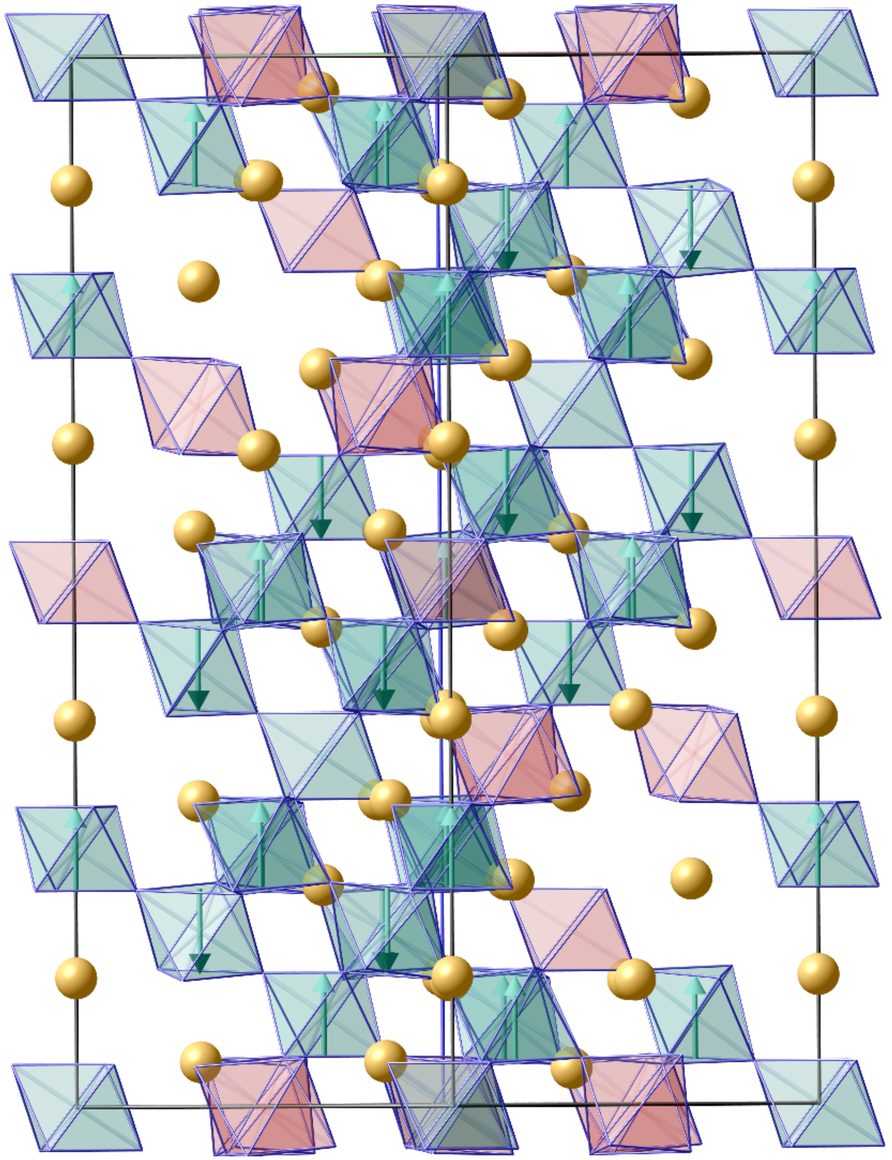
⇒ List of the best incommensurate 10 solutions for 2 satellites

Kx	Ky	Kz	R-factor
0.500000	0.000000	0.500000	0.107554
0.500000	0.500000	0.500000	0.107561
0.500000	0.250000	0.250000	0.150276
0.125000	0.000000	0.000000	0.249546
0.000000	0.125000	0.000000	0.249559
0.125000	0.250000	0.125000	0.320719
0.250000	0.125000	0.250000	0.384592
0.000000	0.000000	1.500000	0.399336
0.250000	0.250000	0.125000	0.430693
0.666670	0.333330	0.666670	0.448512

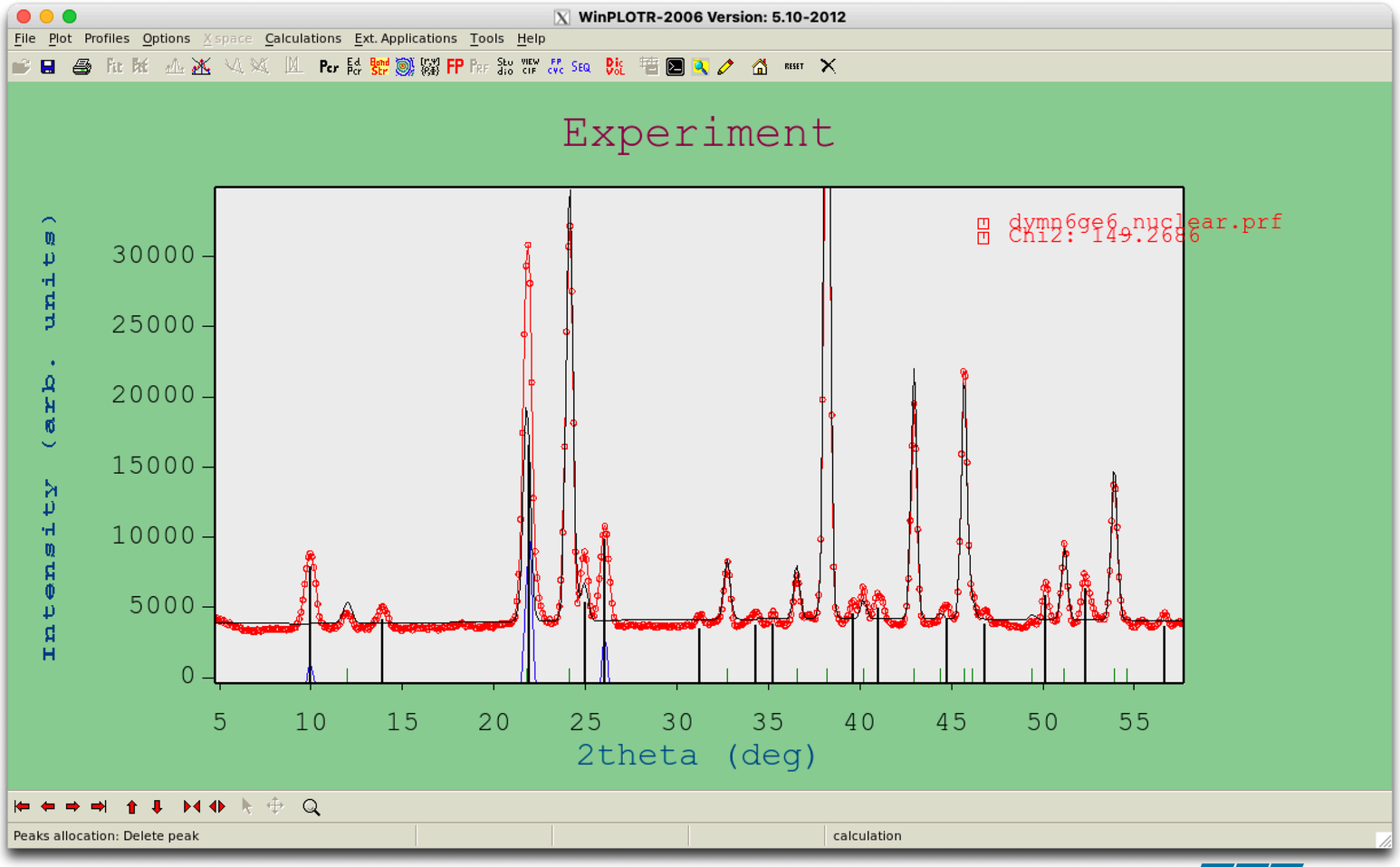
$Sr_4NaRu_3O_{12}$

k_search -> BasIReps etc.

- AF arrangement in triple layers (Ru-⅓Na,⅔Ru - Ru) separated by paramagnetic layer (⅔Na,⅓Ru)
- Triple layers possessing net magnetic moment, arranged AF to each other



$DyMn_6Ge_6$

Difficult case: more than one propagation vector, incommensurability

- Intensity on top of nuclear peaks: $\mathbf{k}_1$=(000)
- k_search only on extra peaks, only three, zero-shift corrected
- Incommensurable…
- Intuition: $\mathbf{k}_2$=(00x)
- Check with Le Bail fit