Magnetoelastic effects in frustrated magnets

Investigation of $LiGa_xIn_{1-x}Cr_4O_8$ "breathing" pyrochlores

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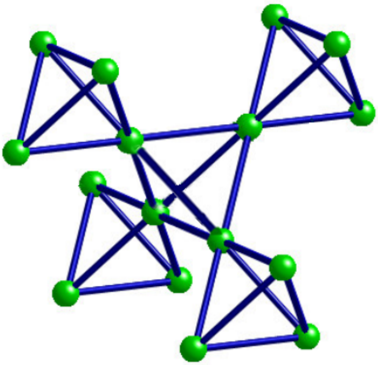
Pyrochlore lattice

Pyrochlore lattice composed of corner-sharing tetrahedra formed by cations.

Most important examples of compounds realizing pyrochlore lattice are:

- pyrochlores $\mathbf{A_2B_2X_7}$, where A and B sites form the tetrahedra
- spinels $\mathbf{AB_2X_4}$, where B site displays a pyrochlore lattice



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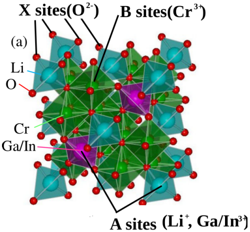
"Breathing" pyrochlore

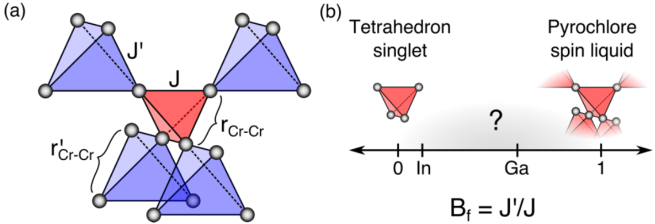
$LiGa_xIn_{1-x}Cr_4O_8$(spinel) **antiferromagnetic** exchange interaction between Cr^{3+} spins($S = \frac{3}{2}$) placed on the corners of tetrahedra. *A* site

populated by Li^+, Ga^{3+} or In^{3+} cations.

Okamoto et al., 2013
Nilsen et al., 2015

Cations are placed alternatively to minimize electrostatic energy.





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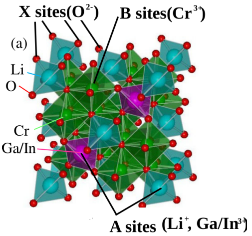
"Breathing" pyrochlore

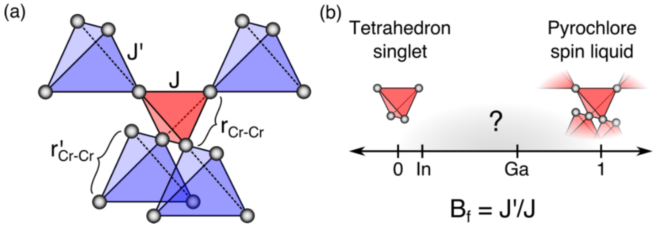
Exchange interactions between Cr^{3+} cations are very sensitive to the distance between the ions.

Composition	r'/r	$B_f = J'/J$
$LiGaCr_4O_8$	1.03	0.6
$LiInCr_4O_8$	1.05	0.1

Okamoto et al., 2013
Nilsen et al., 2015

Cations are placed alternatively to minimize electrostatic energy.





Our main objectives are to determine:

- crystalline structure
- magnetic structure
- structural and magnetic phase transitions
- role of spin-lattice coupling

in $LiGa_xIn_{1-x}Cr_4O_8$ "breathing" pyrochlores.

Powder diffraction(CW)

HRPT(PSI)

Bragg's condition:

$$\lambda = 2d_{hkl} \sin\theta$$

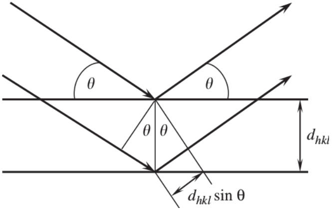


Figure : Source: [1].

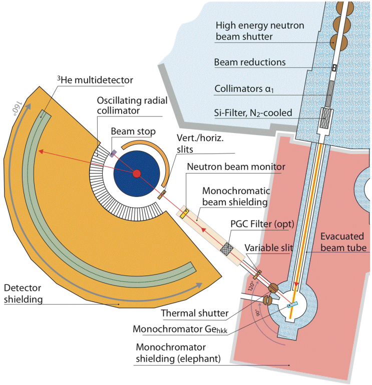


Figure : Layout of HRPT diffractometer at PSI. Source: [2].

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Powder diffraction(CW)

D20(ILL)

Bragg's condition:

$$\lambda = 2d_{hkl}\sin\theta$$

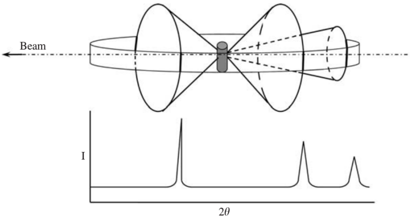

Figure : Source: [1].

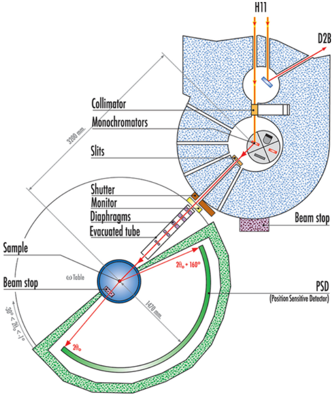

Figure : Layout of D20 diffractometer at ILL. Source: [3].

Powder diffraction(TOF)

WISH(ISIS)

Bragg's condition:

$$d_{hkl} = \frac{ht}{2mL\sin\theta}$$

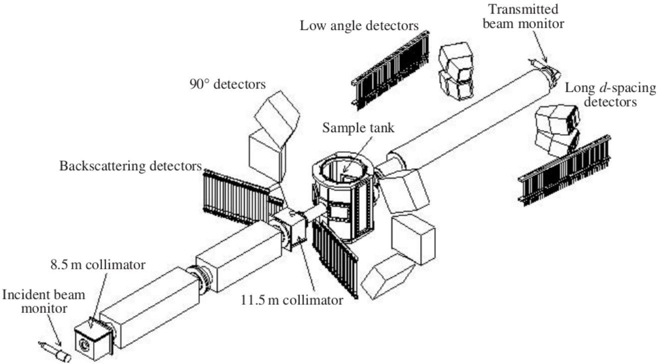

Figure : Layout of typical TOF diffractometer. Source: [1]

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Powder diffractometry

Constant wavelength vs time-of-flight

Constant wavelength

- **peaks are simpler to model**
- incident beam is better characterized
- smipler data storage and reduction
- straightforward absorption and extinction correction
- fine tuning of the resolution during the experiment

Time-of-flight

- whole incoming beam spectrum is utilized
- data are collected to large Q-values(small d-spacings)
- **very high resolution can be achieved by long flight paths**
- **resolution is constant across whole pattern**
- possibility of reducing parasitic scattering from sample environment

Powder diffractometry

Constant wavelength vs time-of-flight - asymmetry

Constant wavelength

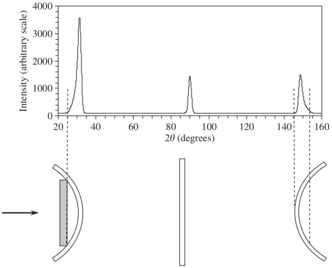

Figure : Asymmetry of Bragg peaks in CW powder diffraction. Source: [1].

$$FWHM^2 = U\tan^2\theta + V\tan\theta + W$$

Time-of-flight

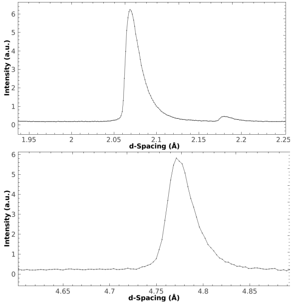

Figure : Shape of the peaks in TOF powder diffraction pattern.

$$\frac{\Delta d}{d} = \left[\Delta\theta^2\cot^2\theta + \frac{\Delta t^2}{t^2} + \frac{\Delta L^2}{L^2}\right]$$

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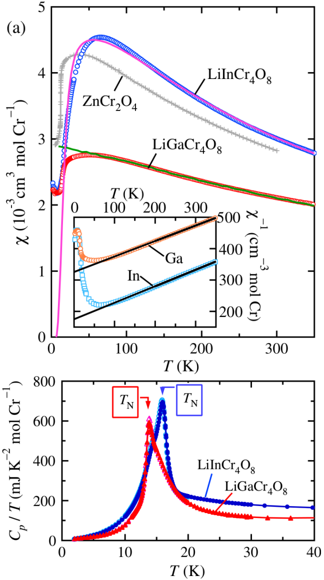
Previous results

Magnetic susceptibility and heat capacity

$LiInCr_4O_8$:

- rapid decrease of χ above 65 K - opening of a spin gap
- energy gap estimated as close to 4.9 meV
- structural phase transition at $\sim$ 16 K
- magnetic phase transition at $\sim$ 14 K

Okamoto et al., 2013

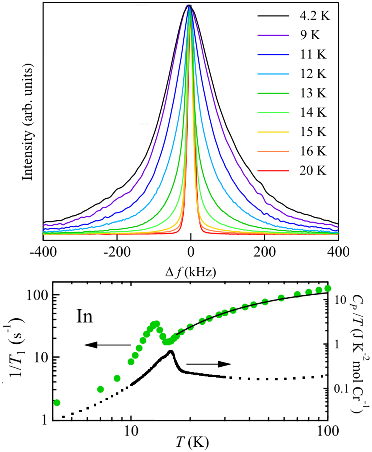


Previous results

NMR

$LiInCr_4O_8$:

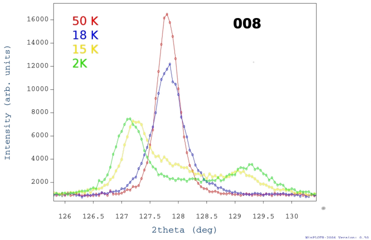
- $1/T_1$ suggest a singlet ground state with a gap of 2.7 meV
- second order AFM transition at 13 K preceded by structural transition releasing frustration

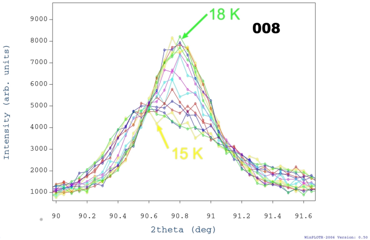


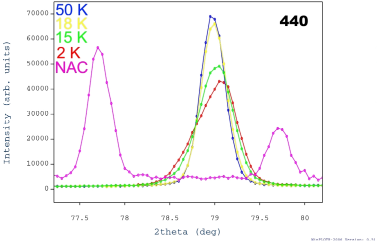
Tanaka et al., 2014

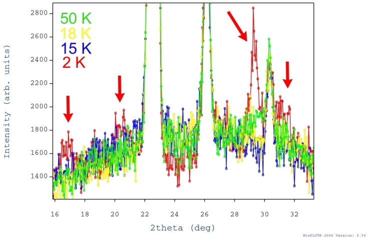
HRPT - PSI(20-24/07/15)

$LiInCr_4O_8$









HRPT - PSI(20-24/07/15)

$LiInCr_4O_8$

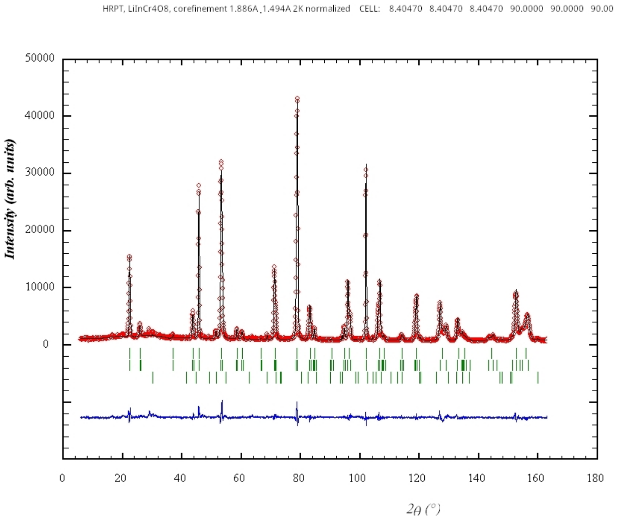


Figure : Rietveld refinement profile of 1.89 Å at 2 K with $\chi^2 = 2.88$.

HRPT - PSI(20-24/07/15)

$LiInCr_4O_8$

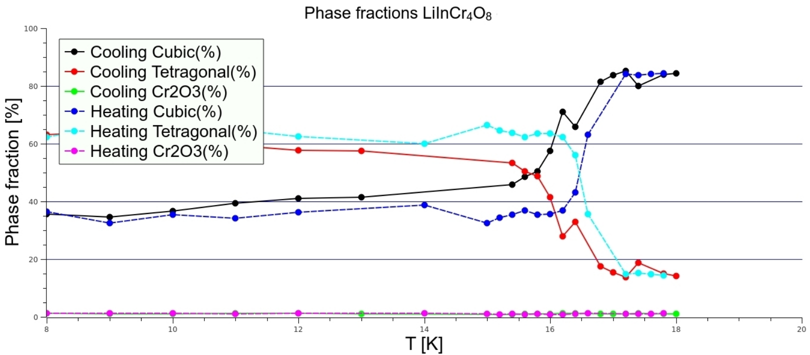


Figure : Temperature dependence of a phase fractions retrieved from Rietveld refinement.

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D20 - ILL(28-29/07/15)

$LiInCr_4O_8$

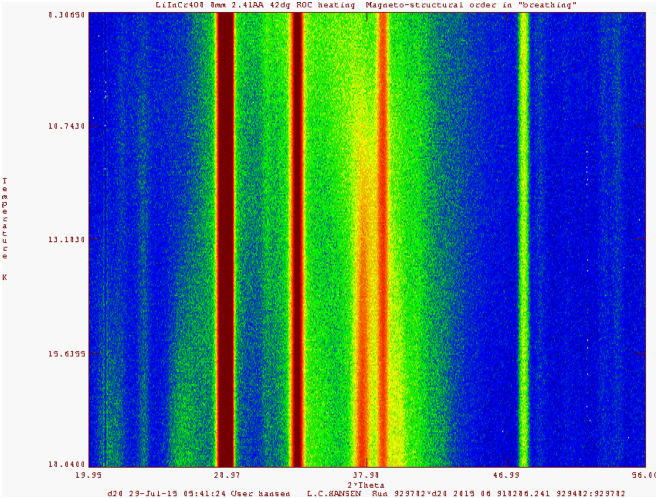

Figure : Temperature dependence of low-angle features from pattern gathered with λ =2.41 Å.

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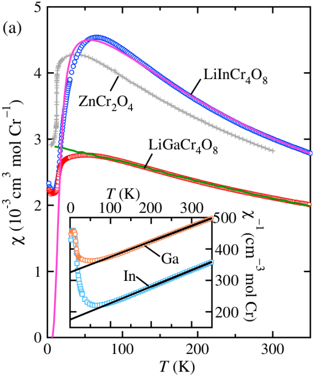
Previous results

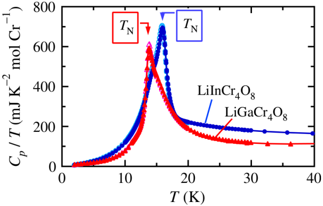
Magnetic susceptibility and heat capacity

$LiGaCr_4O_8$:

- broad peak at $\approx$ 45 K
  -development of AFM short range order
- steep decrease at $\approx$ 14 K
  -long range order sets in
- sharp peak in heat capacity at 13.8 K indicating structural phase transition

Okamoto et al., 2013





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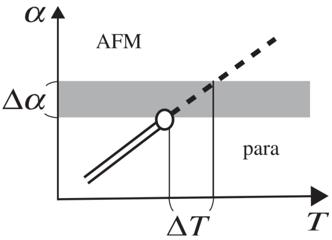
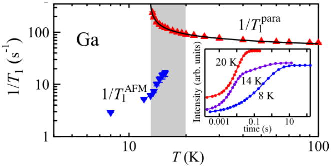
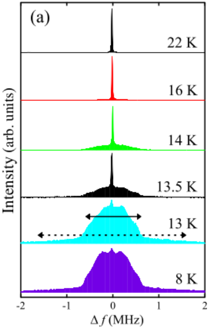
$LiGaCr_4O_8$:

- first order AFM transition
- in the vicinity of tricritical point







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Tanaka et al., 2014

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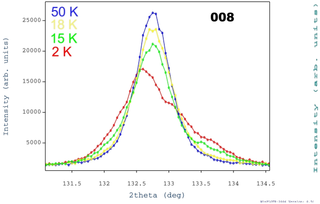
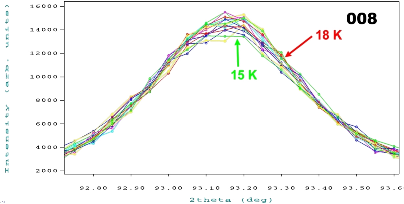
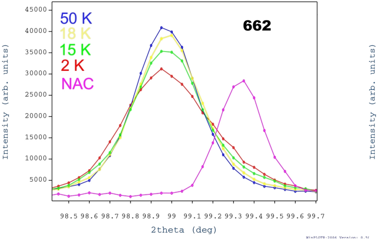
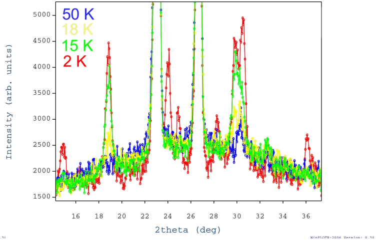
HRPT - PSI(20-24/07/15)

$LiGaCr_4O_8$









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HRPT - PSI(20-24/07/15)

$LiGaCr_4O_8$

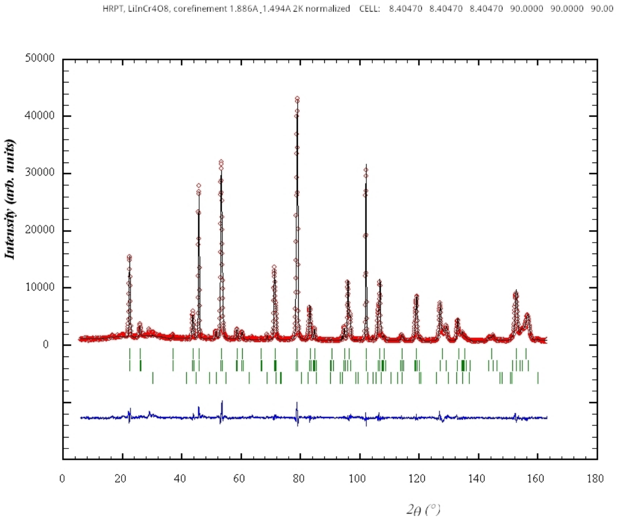

Figure : Rietveld refinement profile of 1.886 Å at 2 K with $\chi^2 = 5.13$.

D20 - ILL(28-29/07/15)

$LiGaCr_4O_8$

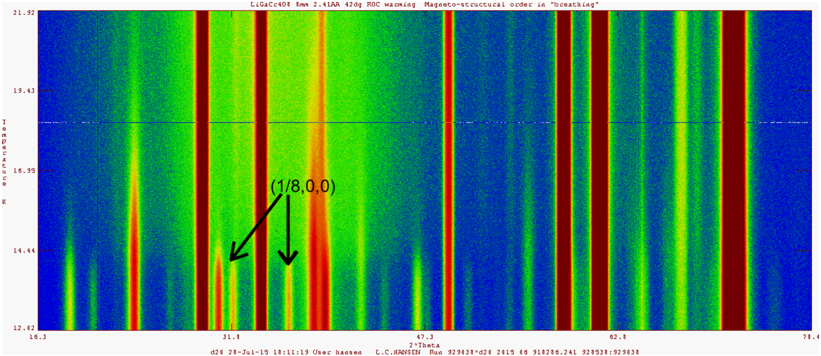

Figure : Temperature dependence of low-angle features from pattern gathered with λ =2.41 Å. Two peaks were indexed as corresponding to (1/8,0,0) k-vector.

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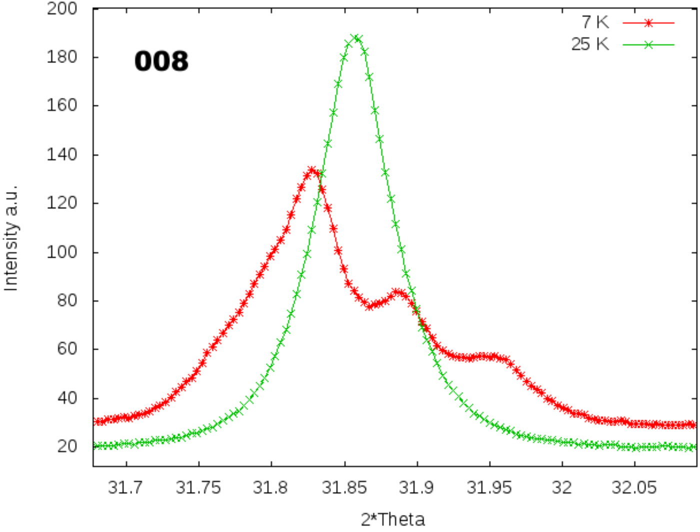
MS BEAMLINE - PSI(27-29/10/15)

$LiGaCr_4O_8$

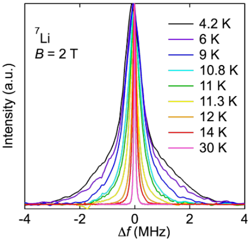


$LiGa_{0.95}In_{0.05}Cr_4O_8$:

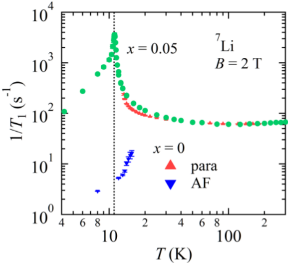
- second order AFM transition

Tanaka et al., not published.

^{7}Li-NMR spectra

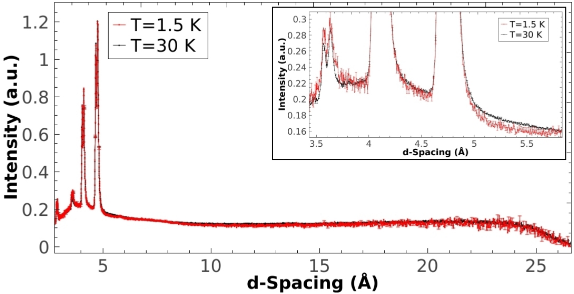


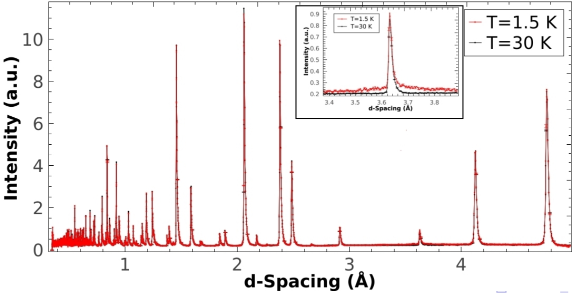
Nuclear Relaxation rate



WISH - ISIS(11-12/10/15)

$LiGa_{0.95}In_{0.05}Cr_4O_8$





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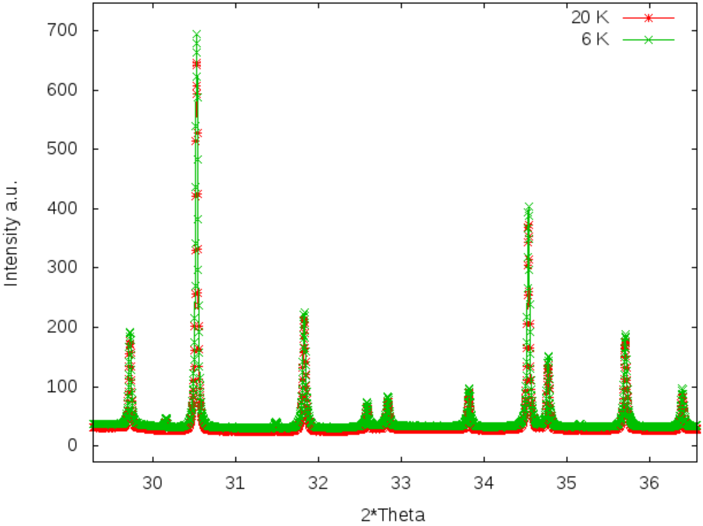
MS BEAMLINE - PSI(27-29/10/15)

$LiGa_{0.95}In_{0.05}Cr_4O_8$



Future perspectives

- determine ordered low T magnetic structure in $LiGaCr_4O_8$
- AC magnetic susceptibility measurements on $LiGa_{0.95}In_{0.05}Cr_4O_8$ sample to confirm presence of spin glass phase
- perform single crystal experiments as soon as such a samples will be available

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References

[1] E.H. Kisi, C.J. Howard, *Applications of neutron powder diffraction*, Oxford University Press, Oxford, 2008.

[2] http://www.psi.ch/

[3] http://www.ill.eu/

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