Neutron diffraction on single crystals under very high pressures in diamond and sapphire anvil cells

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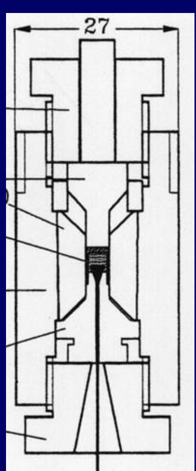
Single-crystal diffrqction studies under extreme contiditions:

pro	*contra*
Precise information on crystal structures, especially low symmetry/distorted ones, or orientation of molecules molecular ordering	**Difficult sample preparation: very small (10-100μ) pre-oriented crystals**
Details on magnetic structures, for example choice between collinear and noncollinear orderings	**Shattering of single crystals under pressure because of nonhydrostaticity or first-order phase transitions**
Possibility to combine pressure with anisotropic conditions such as uniaxial stress or magnetic field	**More expertise required for collecting data**

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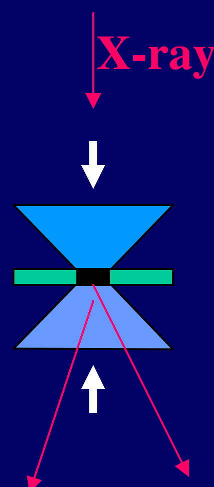
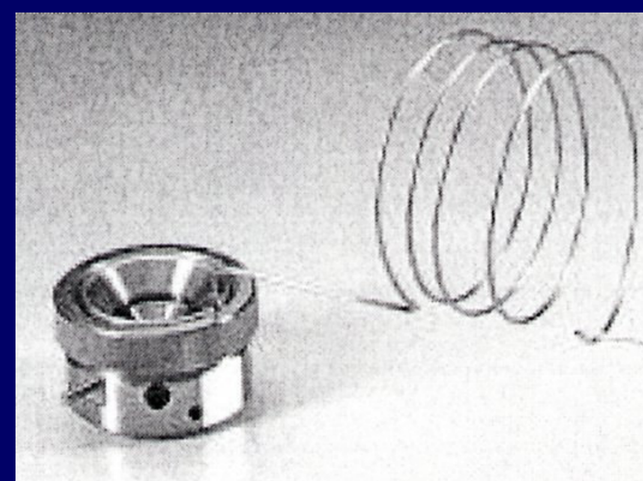

X-ray


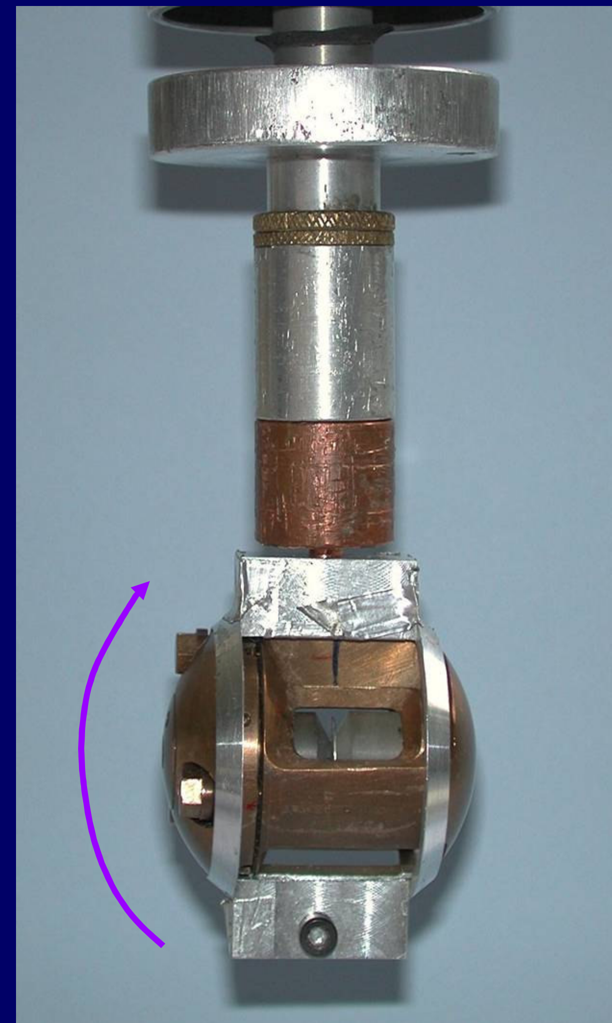
Compact “LLB pressure cells” with sapphire/cBN/moissanite/diamond anvils

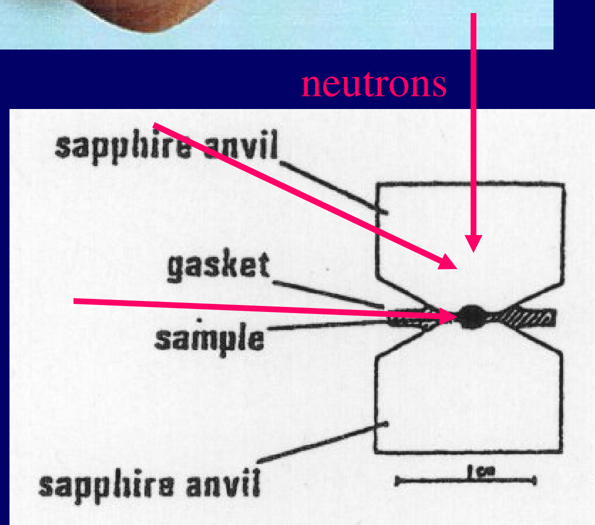
Goncharenko High Press. Research (2004)

5 cm

Max. load= **25 kN** **30 kN** **50 kN** **250 kN**


P=7 GPa,
V= 1 mm3



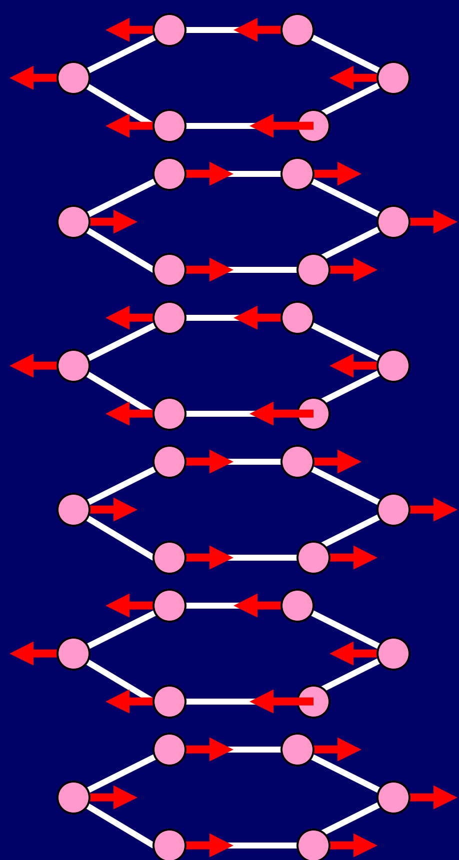

neutrons
sapphire anvil
gasket
sample
sapphire anvil
1 cm

Single-crystals in magnetic studies: provide key information on role of nonhydrostaticity (anistropic pressure component) in high pressure studies

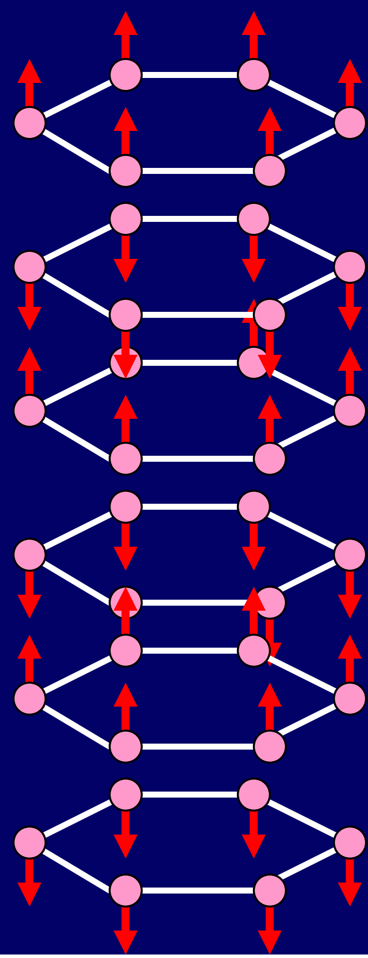
Morin transition in hematite α-Fe_2O_3

Orientational magnetic transition at $T=T_M$ ~260K (after Shull, Wollan, 1951)

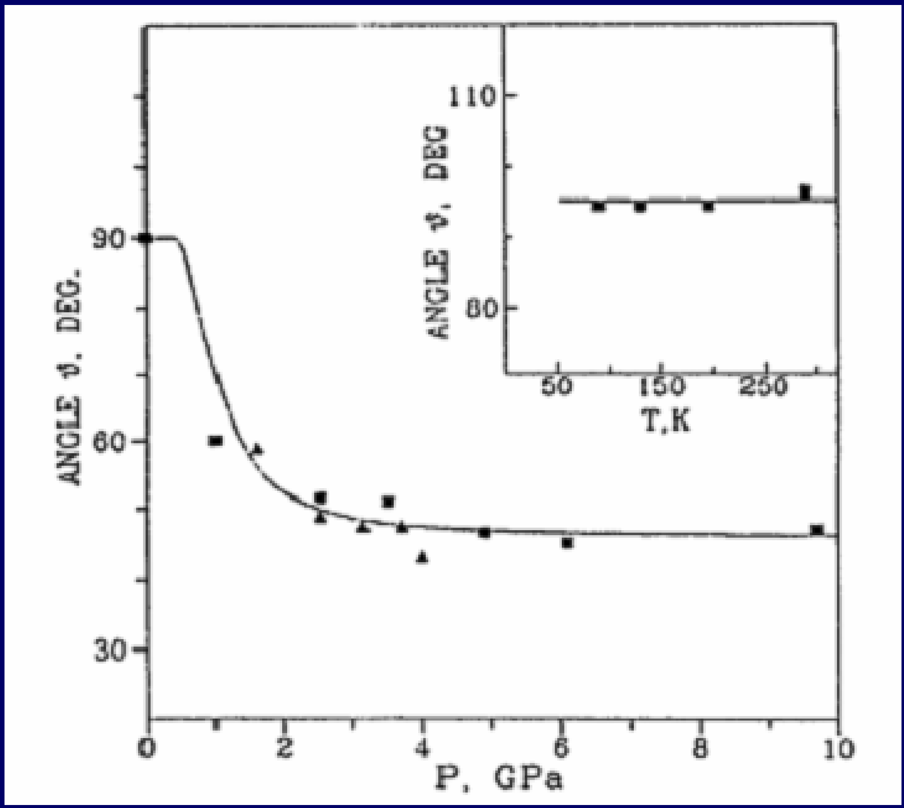
$T>T_M$

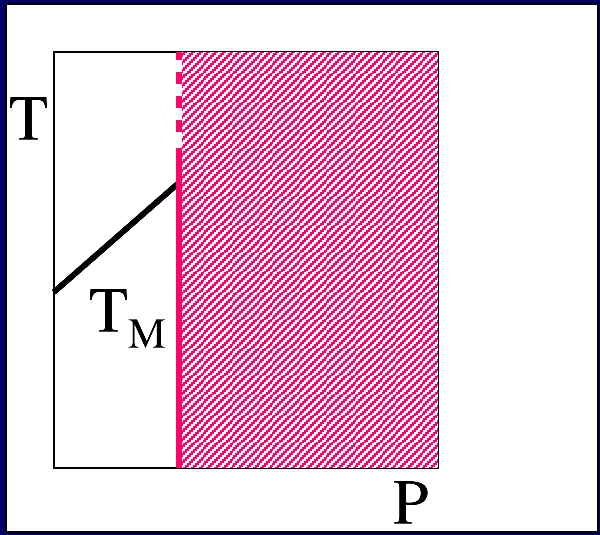


$T<T_M$



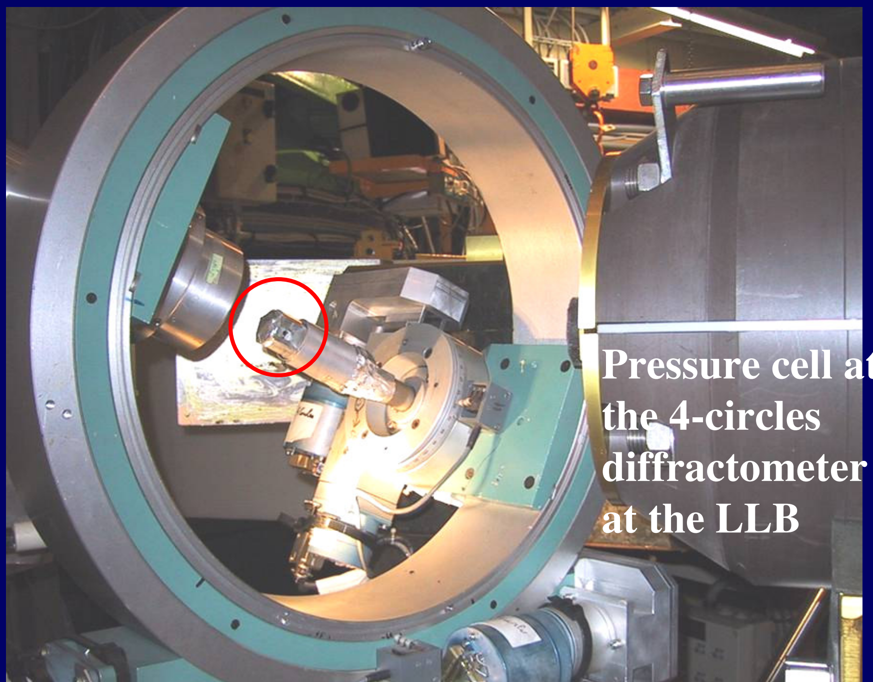
High Pressure Powder Neutron Studies:
Worlton et al., 1968 up to 4 GPa
Goncharenko et al. 1995 up to 10 GPa

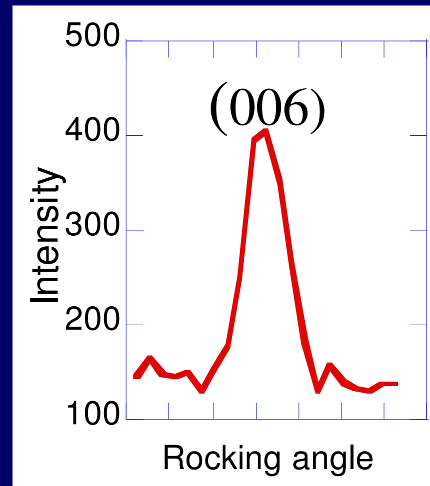


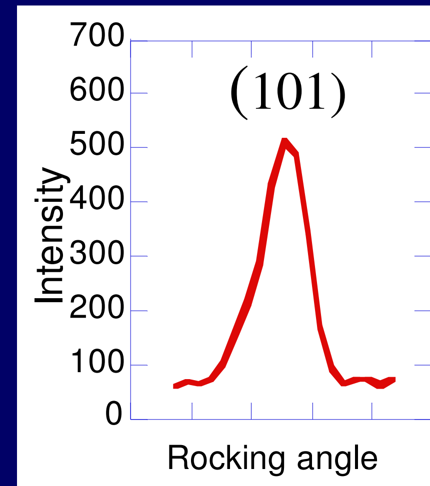


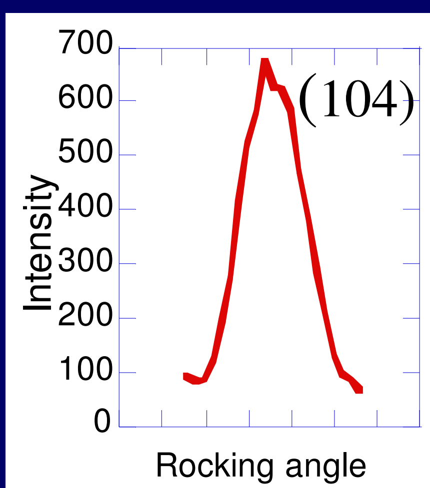
At low pressures, T_M seems to increase, but at higher pressures magnetic moments freeze in some intermediate direction

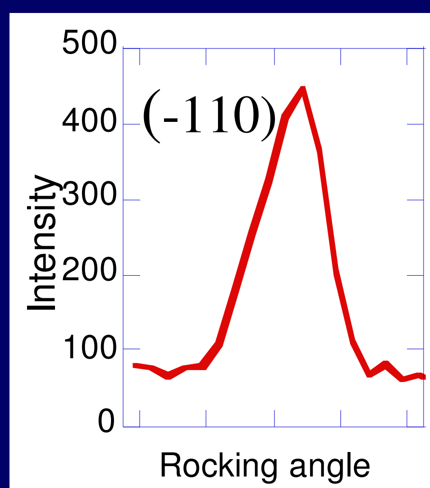
2006: single crystal study in hydrostatic conditions. About 30 magnetic reflections measured at P=4 GPa.

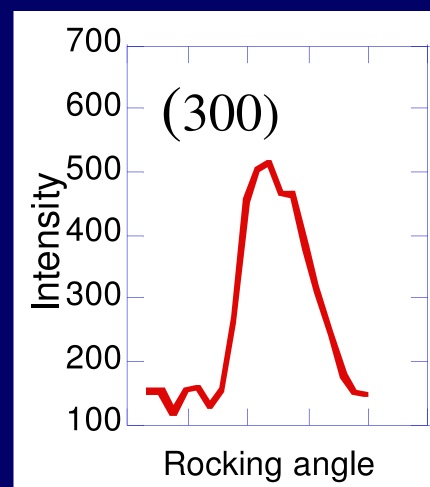


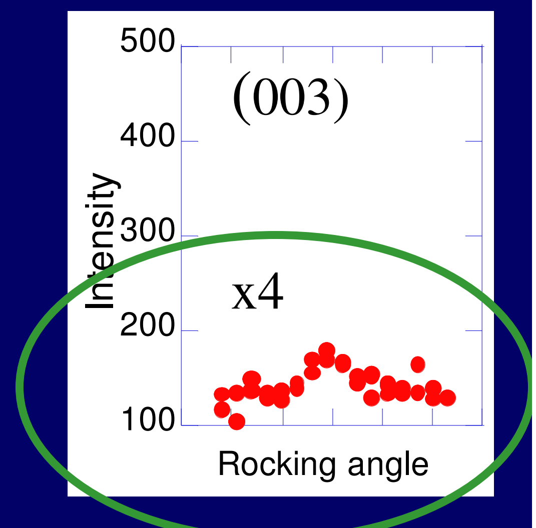




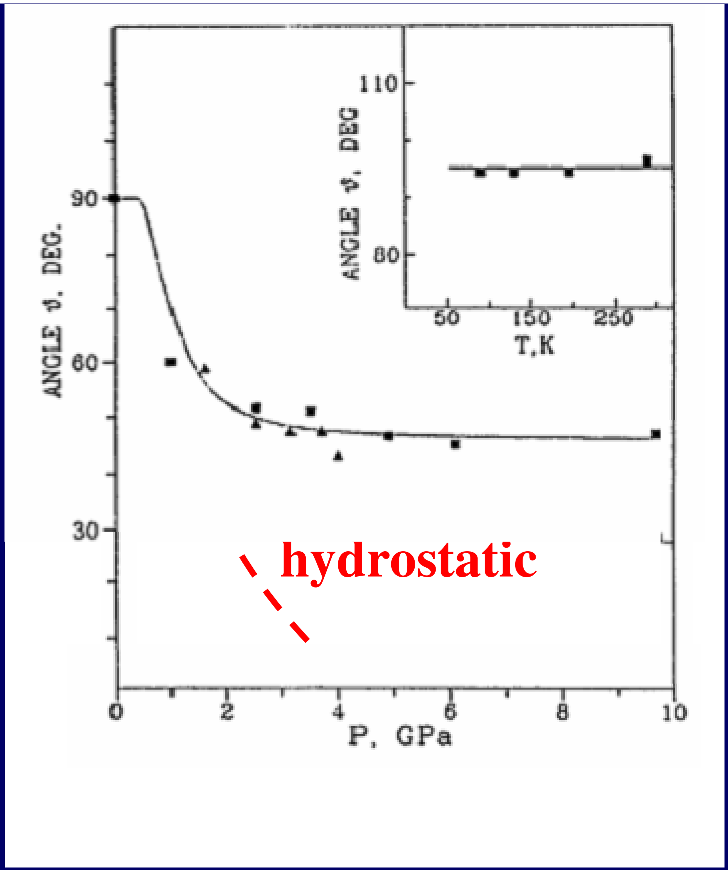








The magnetic moments along the rhombohedral axis within +/- 5°



In non-hydrostatic conditions the Morin transition is controlled by uniaxial stress; hydrostatic pressure allows to complete the transition

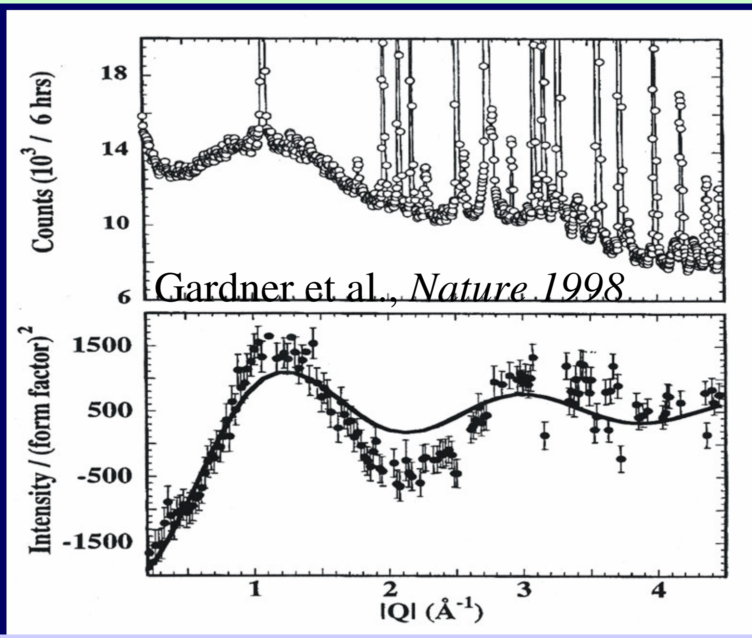
$$F_{magn} = \Sigma J_{(R-R_i)} SS_i + \Sigma \delta J_{(R-R_i)} \delta R \cdot SS_i + \mu H$$

isotropic Heisenberg part; allows to study microscopic origins of magnetic interactions

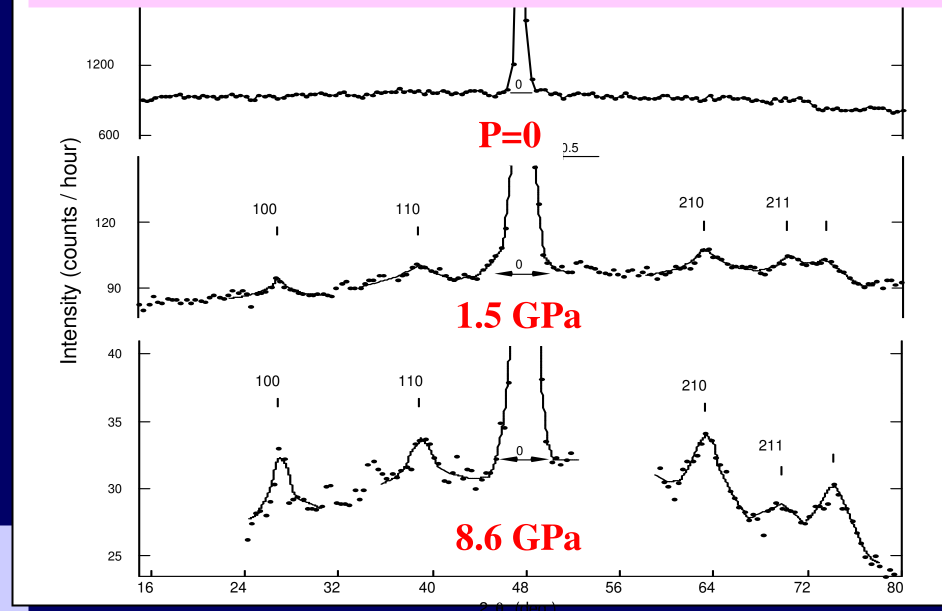
anisotropic term induced by stress; allows to study role of topology in magnetic interactions

anisotropic term induced by field; allows to estimate energy scale of ferro- or antiferro- interactions

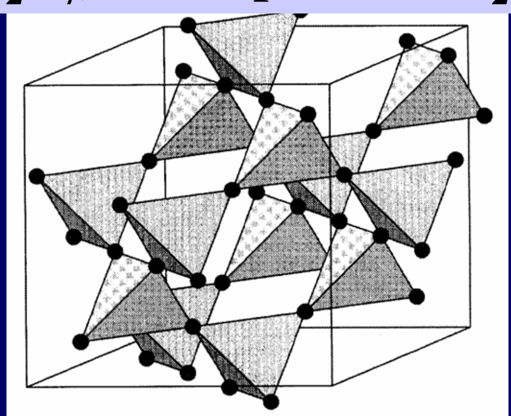
$Tb_2Ti_2O_7$: a quantum spin liquid down to T=0.1 K



Ppessure-induced crystallization of the spin-liquid, *Nature, 2002*

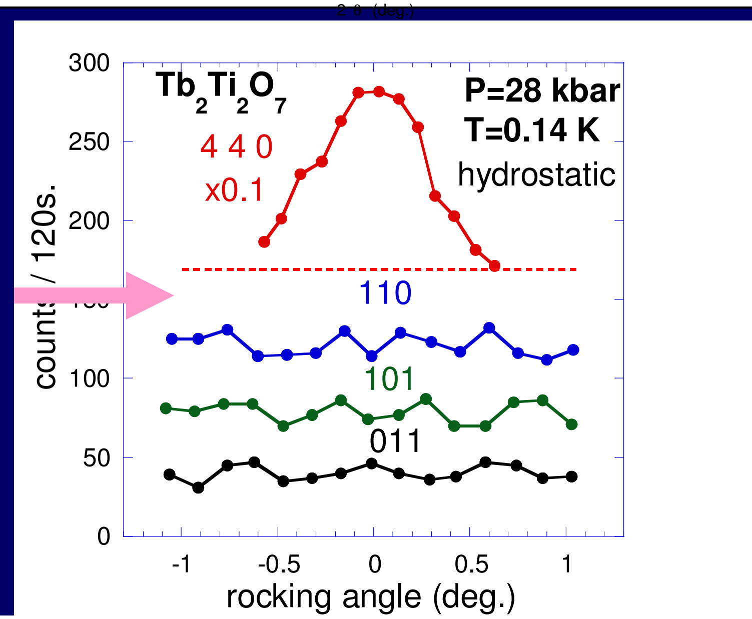


corener-shared lattices: pyrochlore $A_2B_2X_7$, Laves phases AB_2

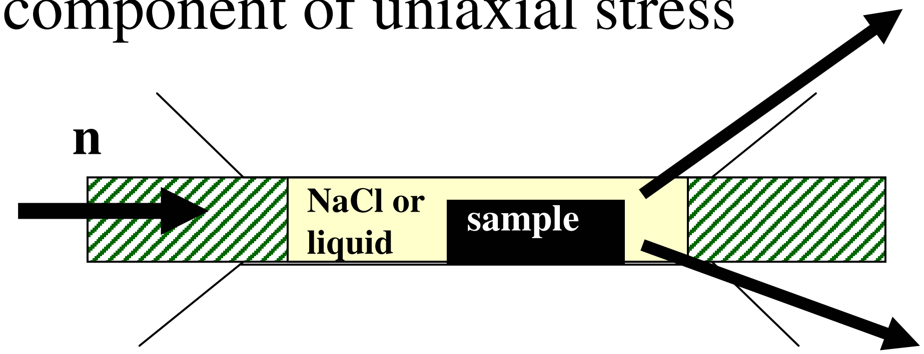


degenerated regarding first-neighbor AF interactions

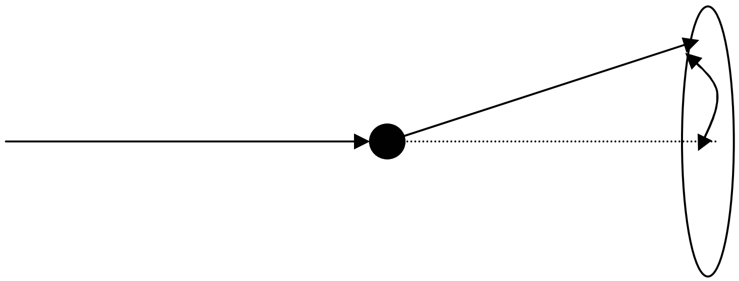
hydrostatic pressure on a single crystal



Measurements with a controlled component of uniaxial stress

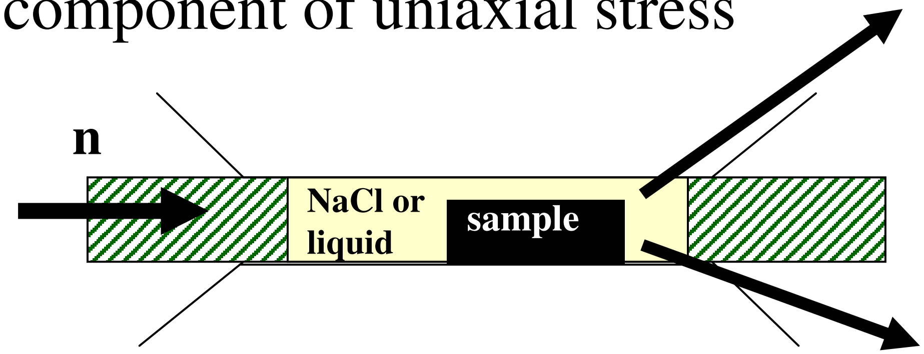


Measurement of anisotropic stress by scanning the diffraction cone from NaCl



lifting counter single-crystal diffractometer 6T2
λ=0.09-0.24 nm

Measurements with a controlled component of uniaxial stress



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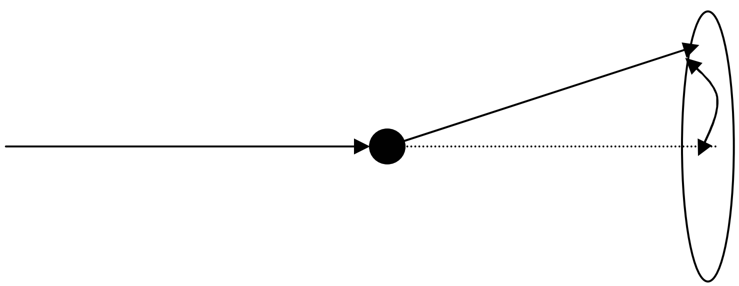
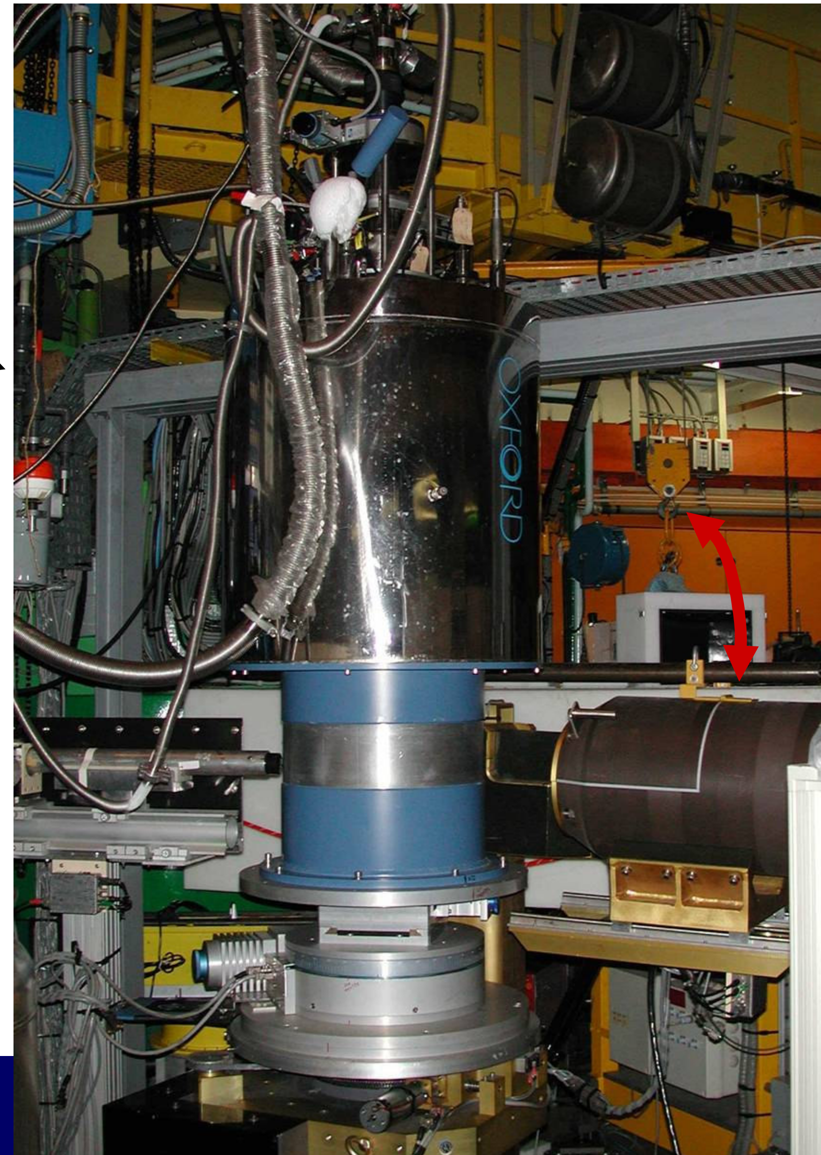
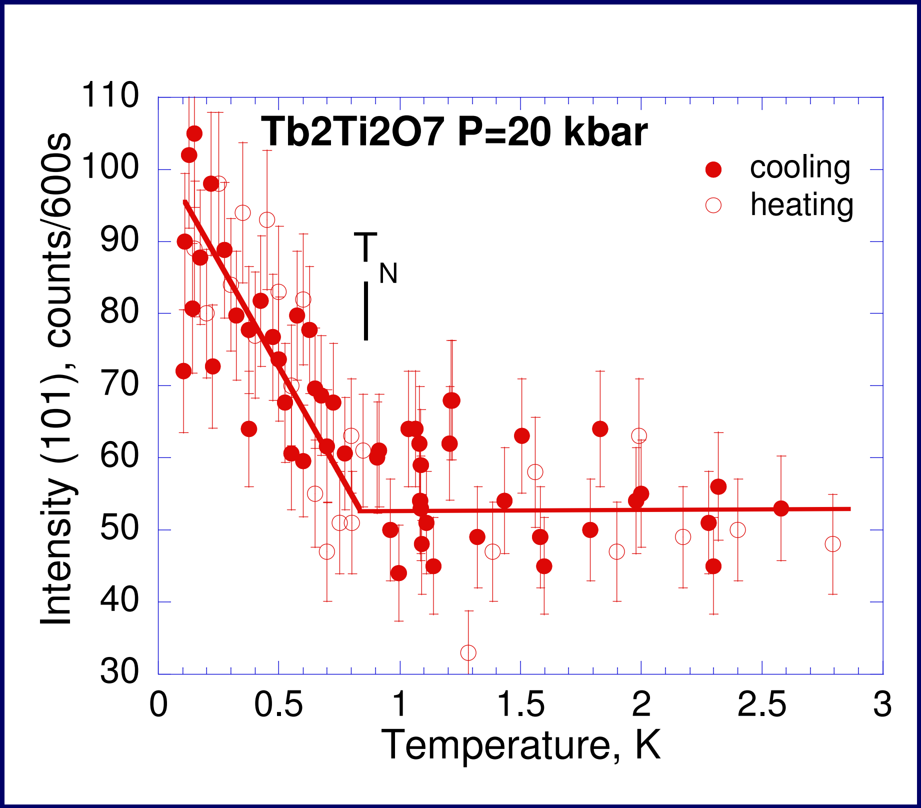
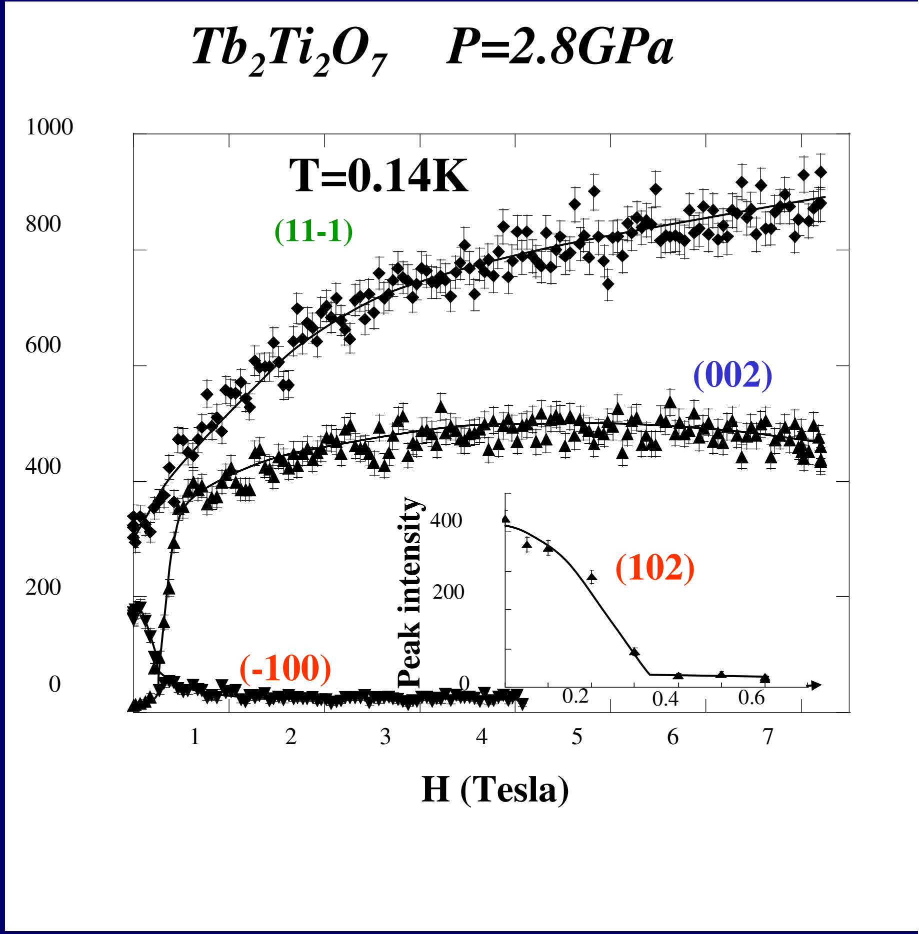




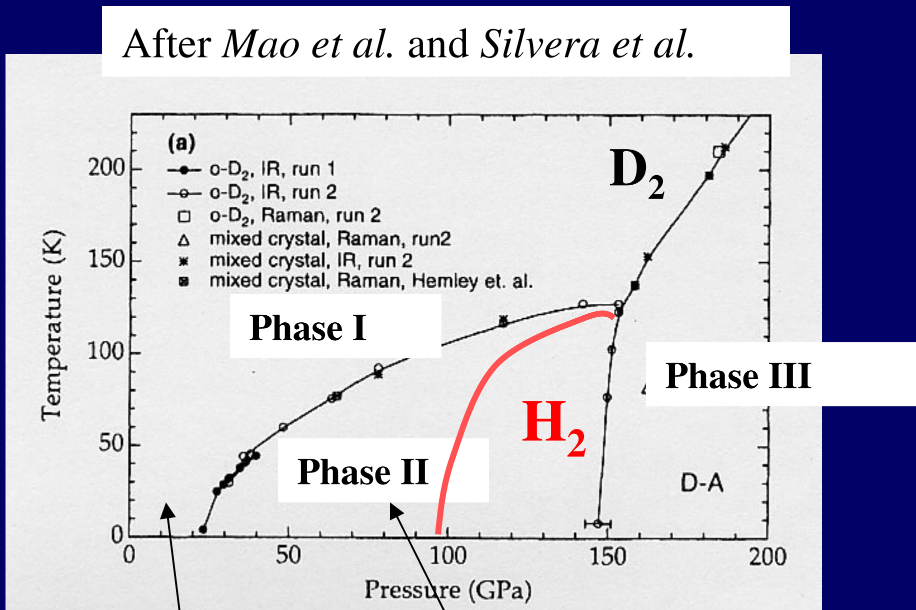
Pressure (2 GPa) *and* anisotropic stress (0.3 GPa):
Crystallization of the spin liquid!
Long range magnetic order (T_N = 0.8 K)

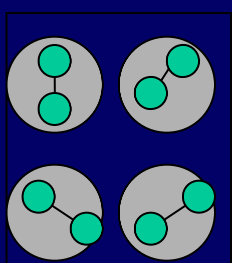


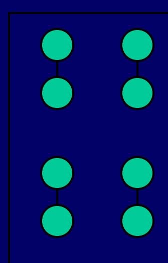


Mirebeau, Goncharenko, Revkolevski et al, *PRL 2004*

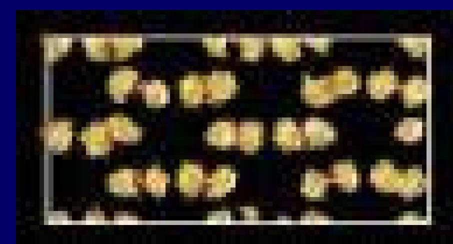
Simple molecular solids H_2, D_2 under pressure. Complementary neutron and X-ray studies.







Ab-initio calculations for phqs II:
Johnson&Ashcroft, Nature 1997,2000
Kitamura et al., Nature 2000
Kohanof et al., PRL 1997
Different predictions, no experimental proof!

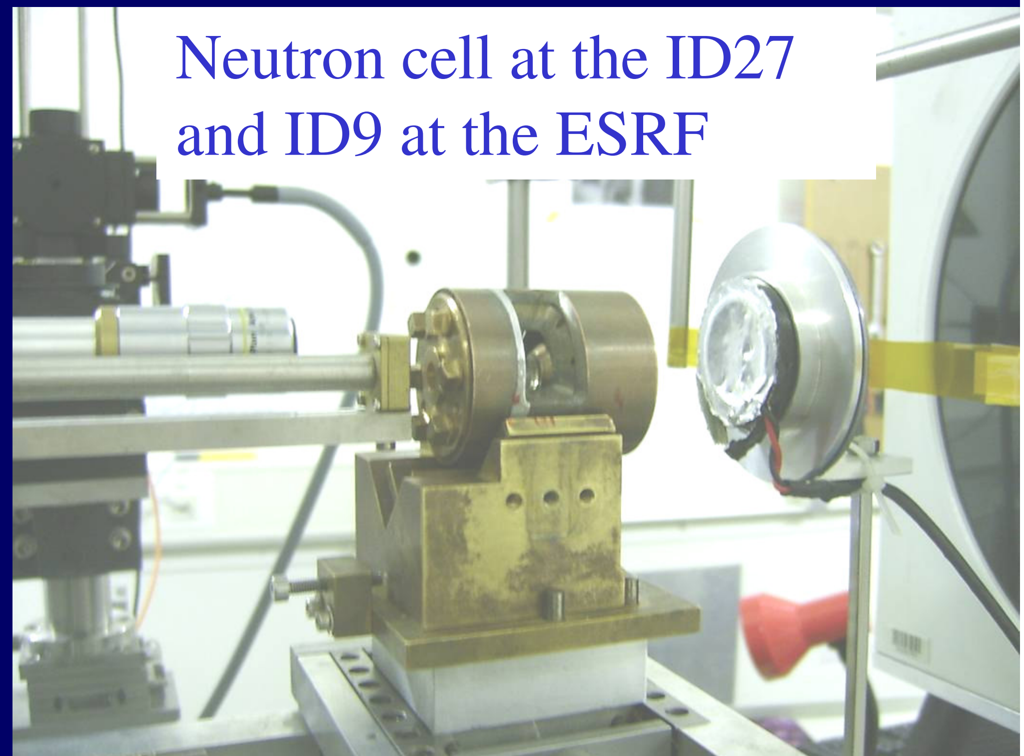


…of course, X-ray or neutron diffraction studies at a level sufficient to determine not only unit-cell parameters but also the contents of the unit cell would provide the ultimate test of the structure… (Edwards & Ashcroft, *Nature 1997*)

Combined X-ray/neutron study *on the same sample in the same thermodynamical conditions :*

the most powerful method to study crystal structures in extreme conditions?

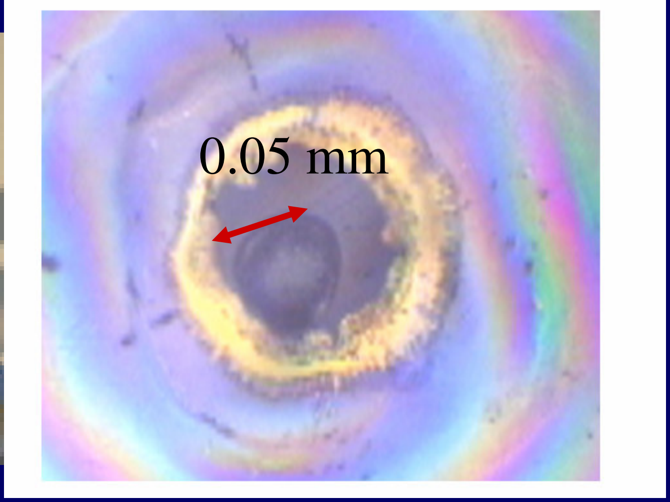
Neutron and X-ray studies on the *same sample* in the *same P-T conditions*



New LLB "hybrid" cell: membrane and screw drive; axial and radial scattering geometries; compatible with neutron and X-ray instrumentations

Neutron and X-ray diffraction study of the broken symmetry phase transition in solid deuterium

Goncharenko&Loubeyre, *Nature*, 2005



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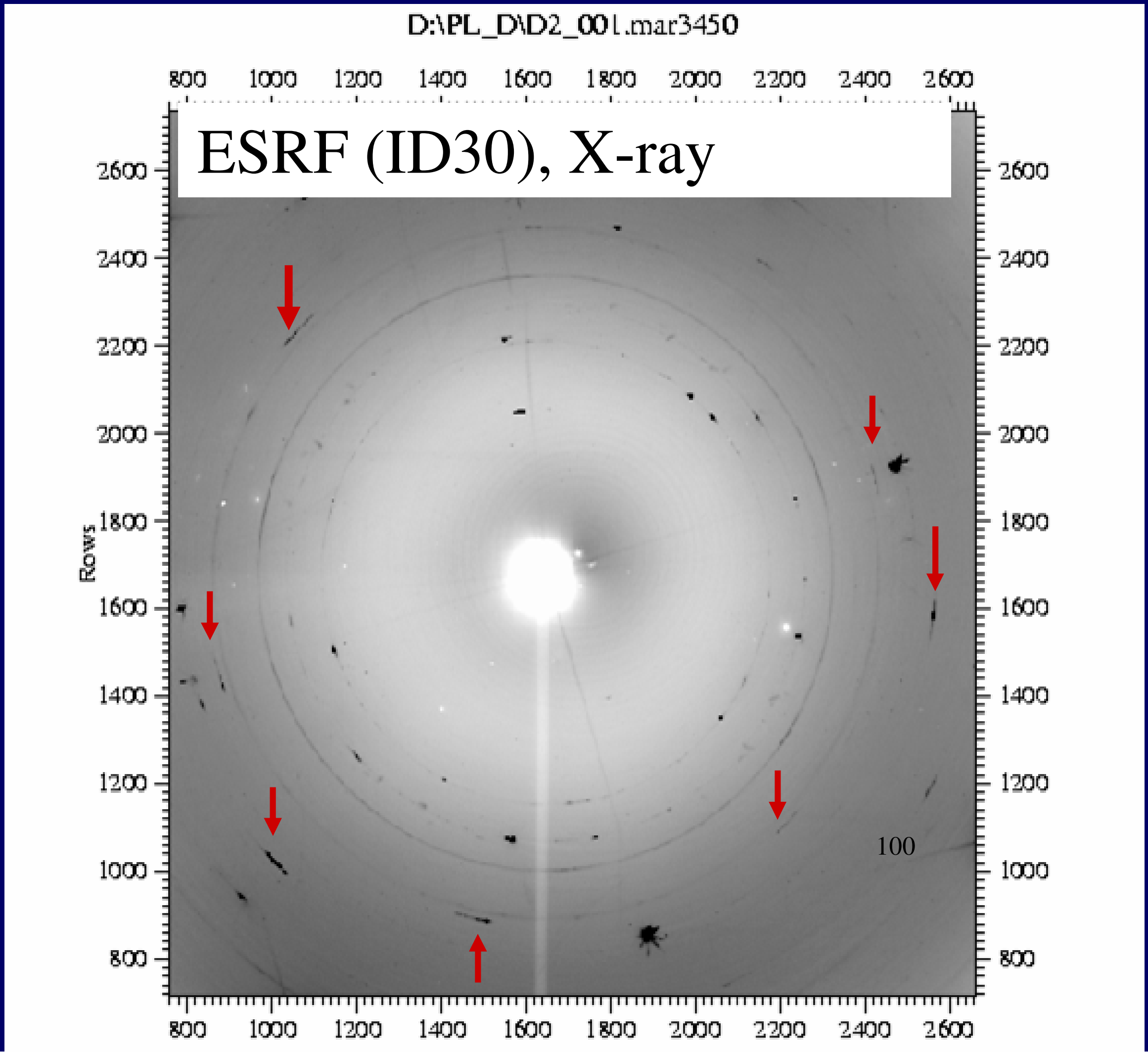

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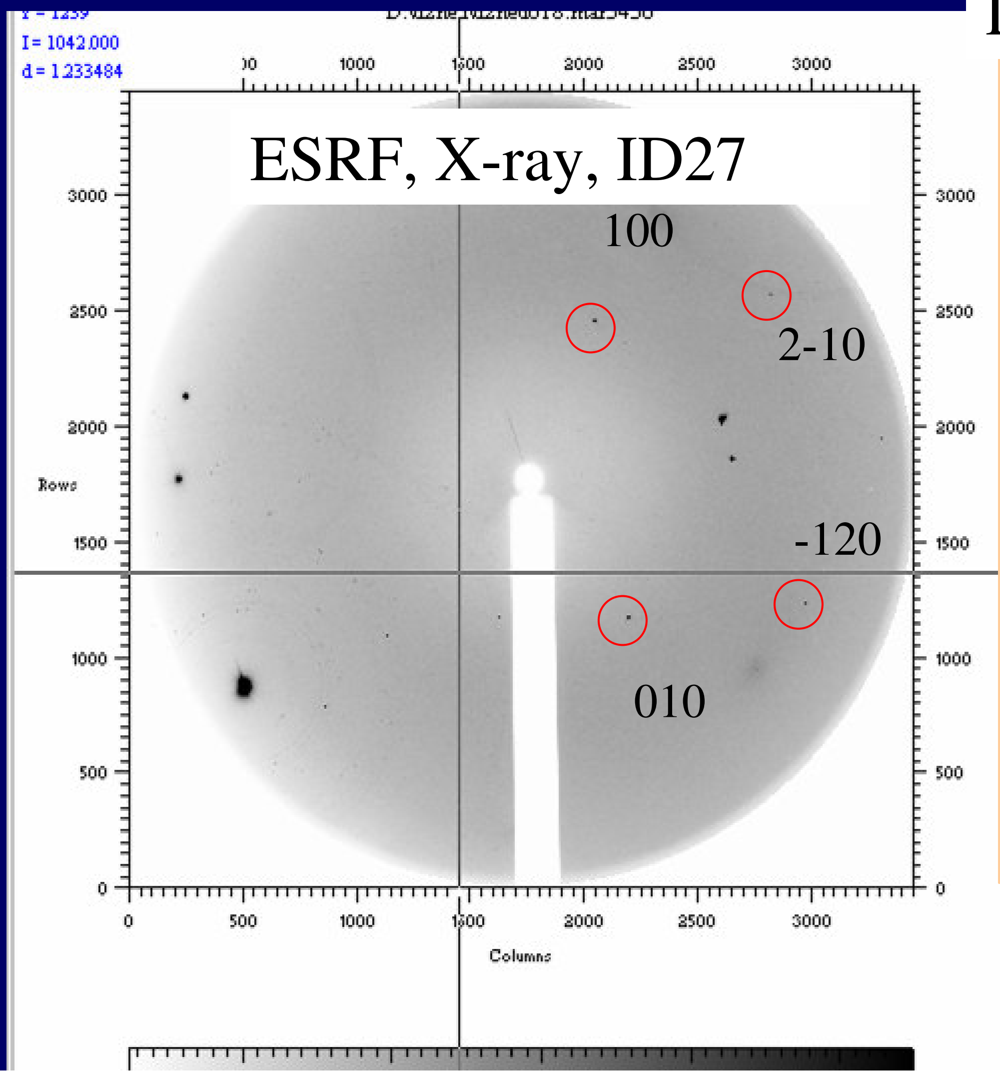
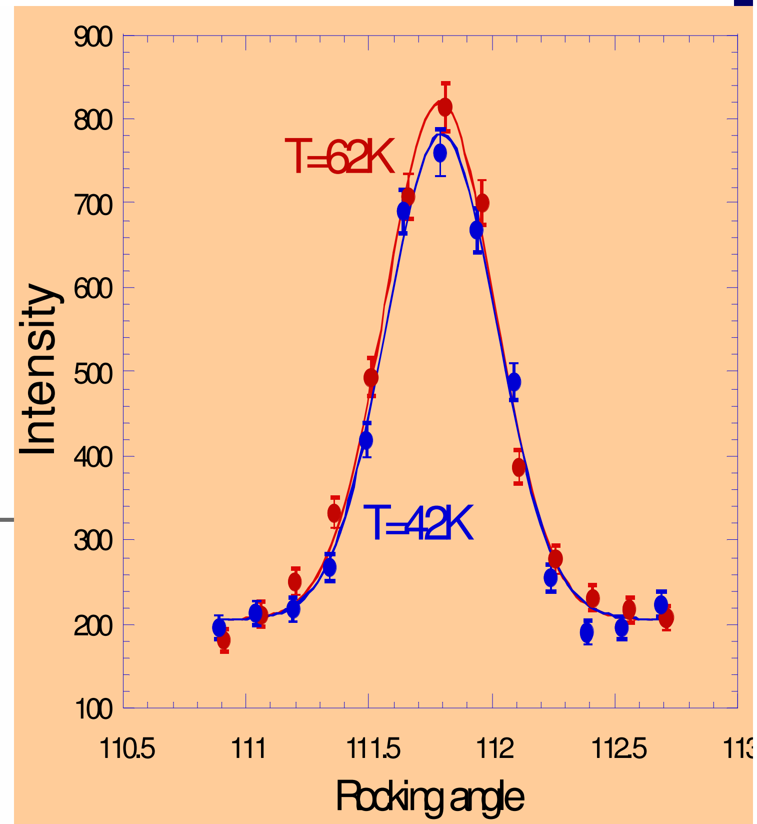
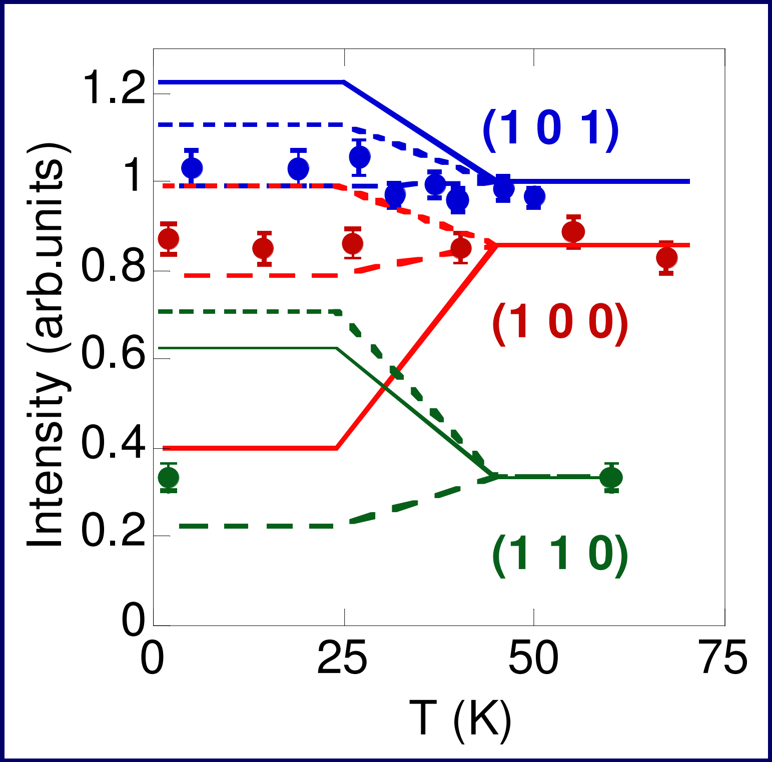
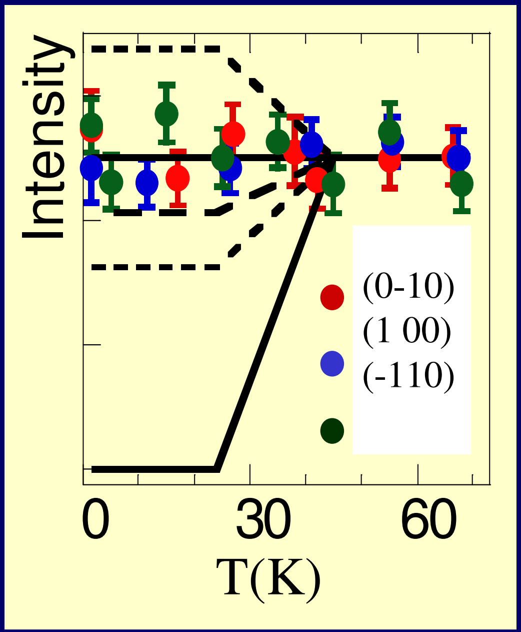


LLB, neutrons, P=38 GPa



A “quadrupole” order, similar to what was found in metastable f.c.c. orhto-para mixtures at P=0?





The variations in intensities trough the transition are much weaker than those predicted by ab initio calculations

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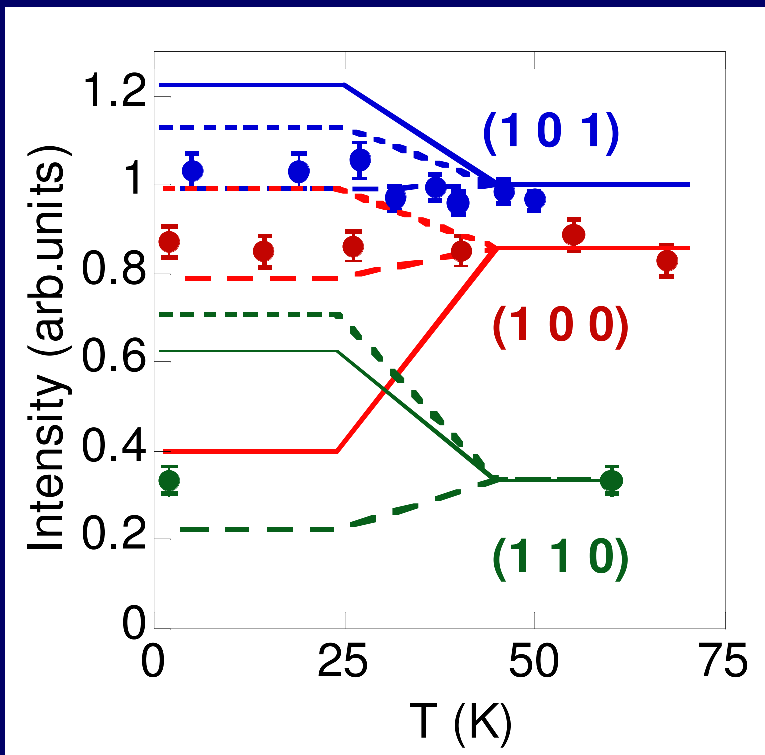

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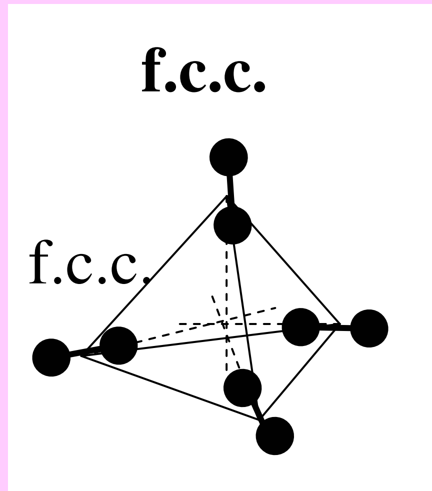
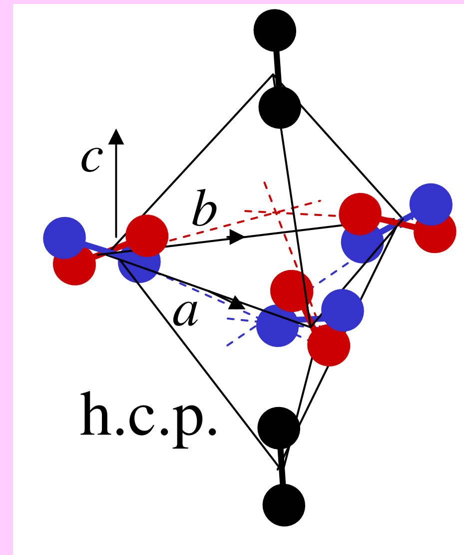
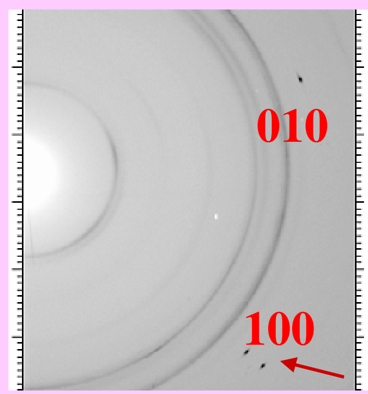
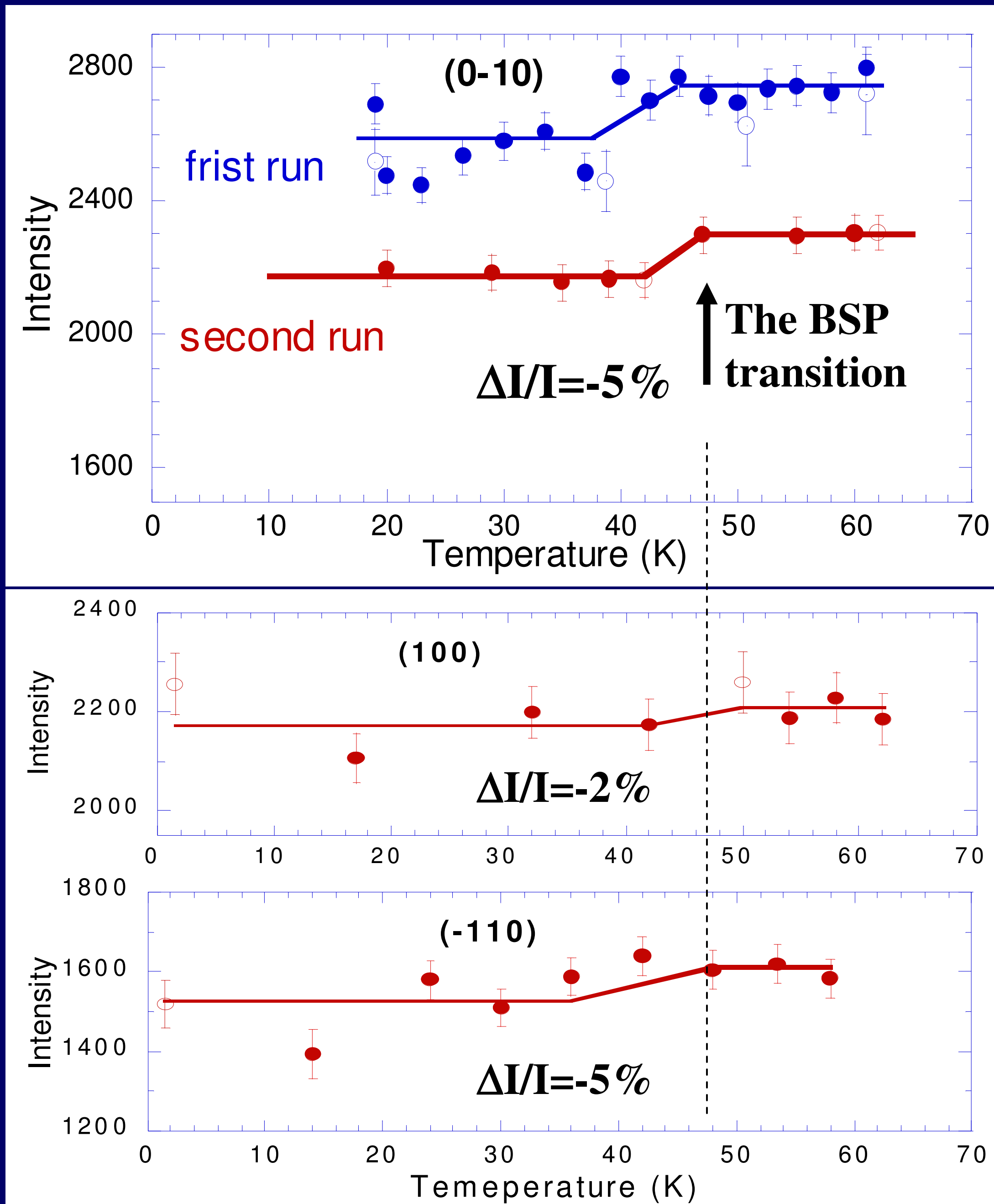


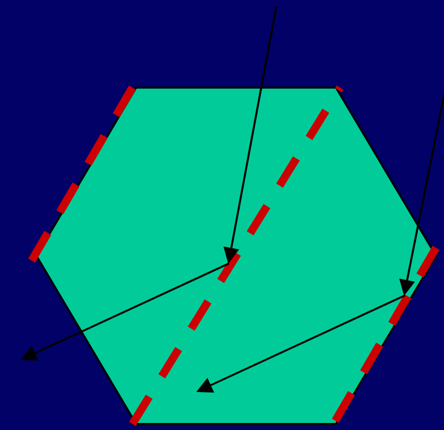


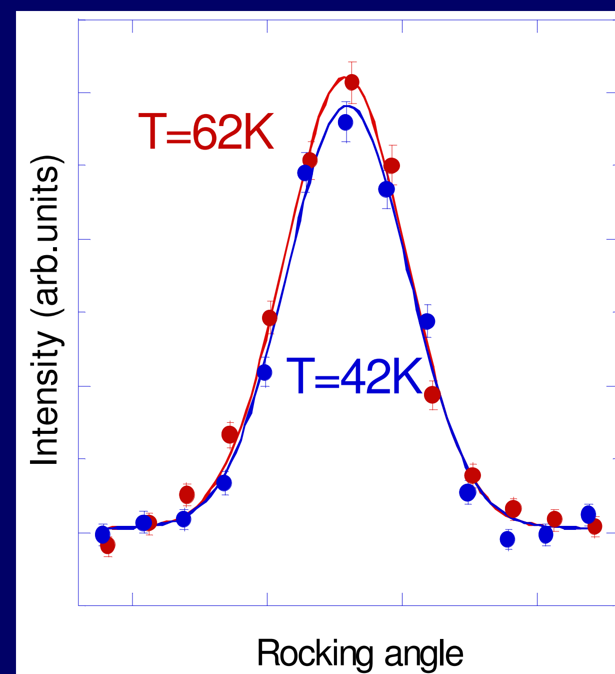


Topoligical frustration of the Pa3 type order in the h.c.p. lattice leads to an incommensurate modulation in the *a-b* plane?

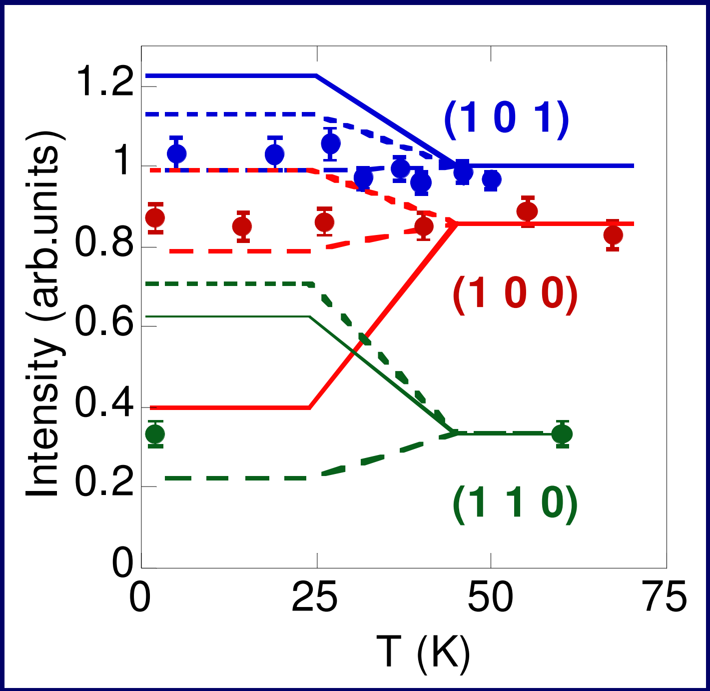
(100), (0-10), (-110) :
equivalent reflections in the P3 cell

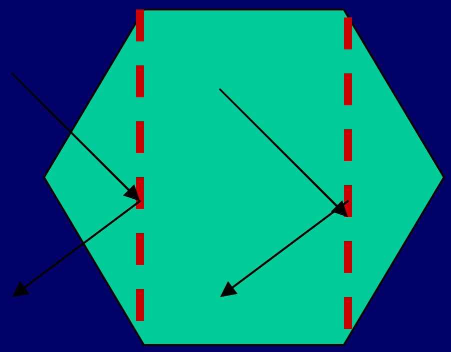






(-120), (2-10), (-1-10) : weaker intensities, but stronger sensitivity to the BSP transition





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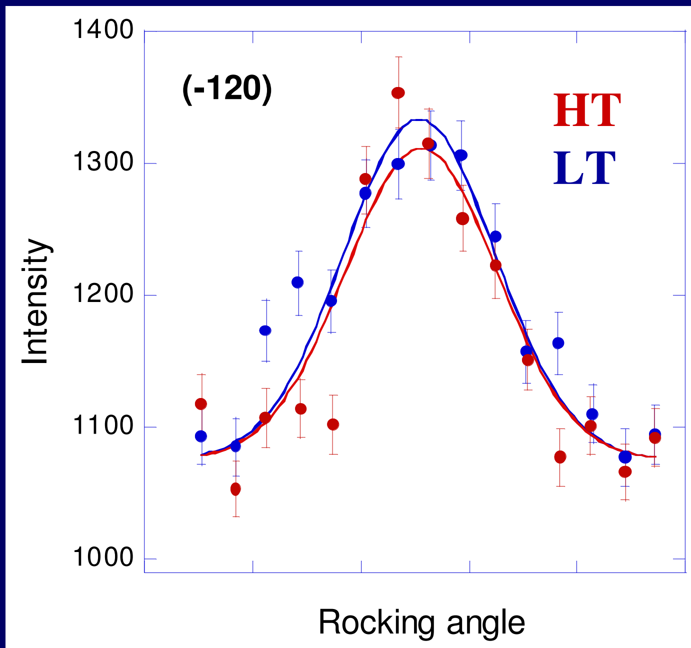
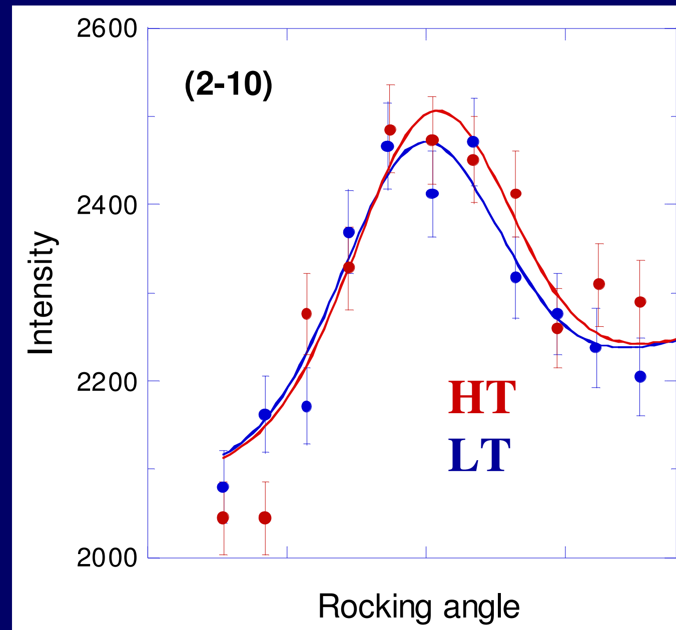
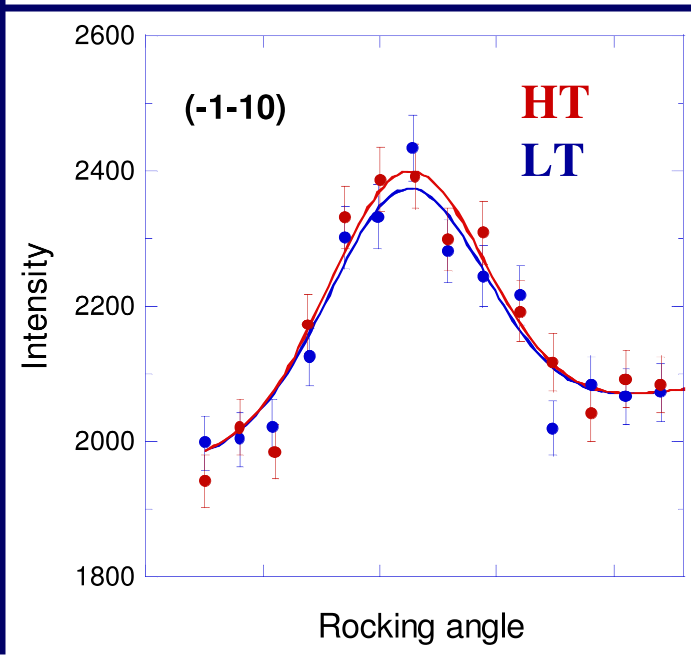
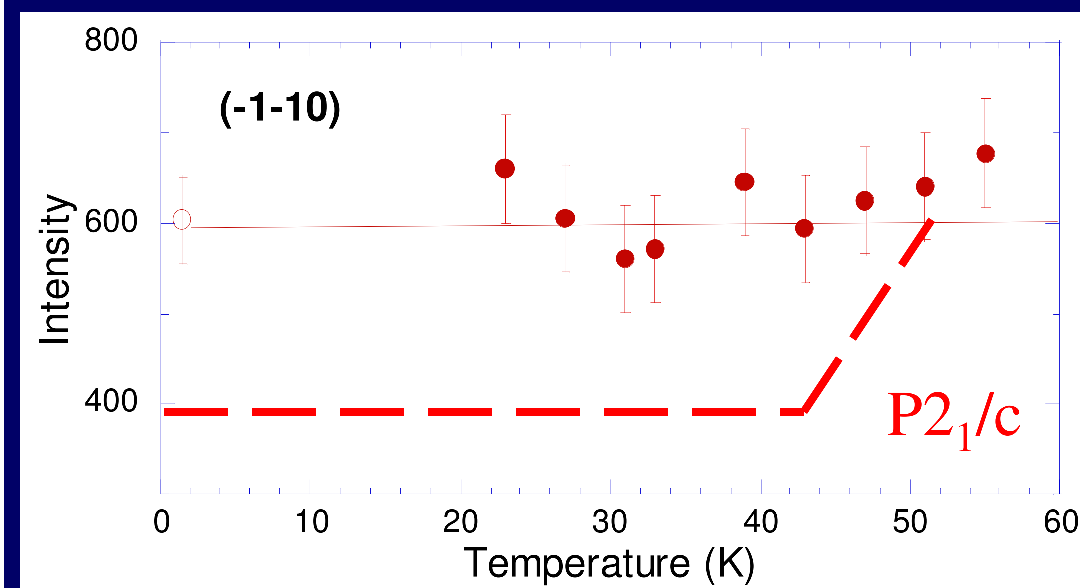


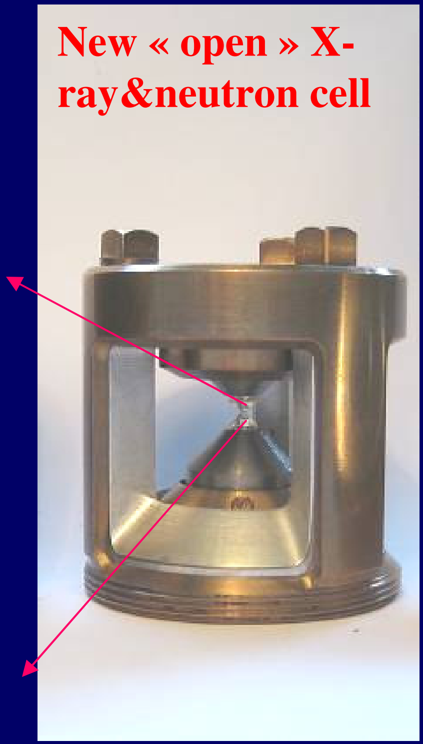






New « open geometry » pressure cells compatible with X-rays and neutrons

High Press. Research, 2007



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J.-M. Mignot

M. Mezouar

M. Hanfland