X-Ray Diffraction Studies of Strongly Correlated Systems in High Magnetic Fields: Current and Future Research Opportunities

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- Research projects (examples).

- Future research opportunities (strongly correlated systems, a biased and personal point of view).

- Existing facilities. Diffraction in high magnetic field at the NSLS.

- What kind of an experimental facility is needed. An invitation to discussion.

A very basic comment.

Magnetic field is one of the few fundamental thermodynamic variables, together with temperature and pressure.

Some statistics: More than 10% of all the recent *Phys. Rev. Lett.* and *Phys. Rev. B* publications have something to do with effects of a magnetic field.

X-ray diffraction is an invaluable tool to study magnetic field effects in matter.

-direct structural changes in an applied field

-magnetic x-ray scattering

-indirect coupling via charge/orbital ordering effects. Resonant scattering.

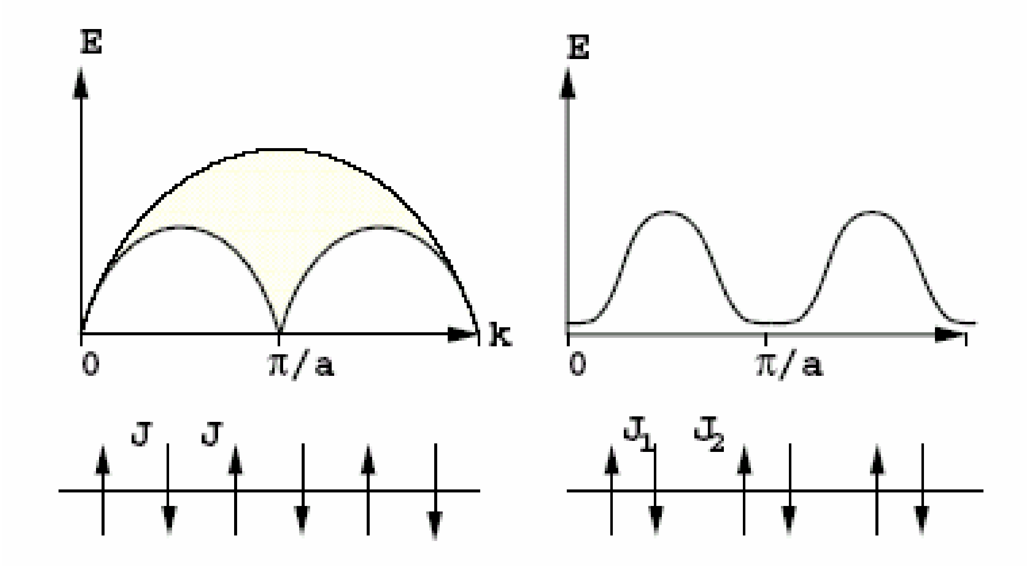
-effects on lattice dynamics and electronic excitations (inelastic scattering)

-more on this later

Research Projects (examples)

1. Model Systems: 1D Quantum Magnets

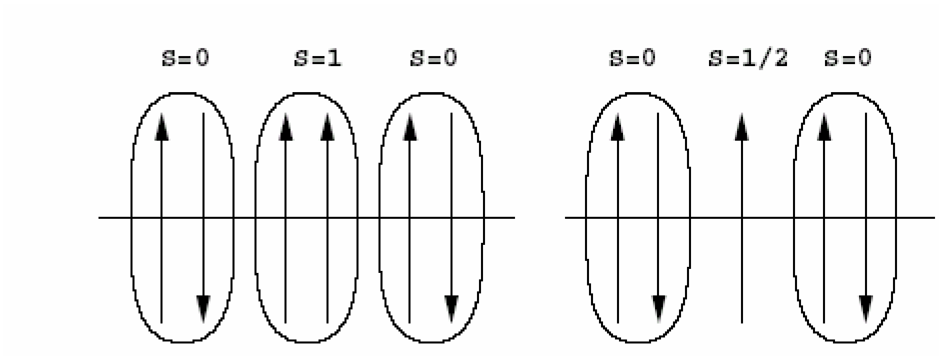
Spin-Peierls Compound $CuGeO_3$



1D chains of spin 1/2 on a deformable 3D lattice

$T>T_{SP}$ $T<T_{SP}$

Effects of a Magnetic Field

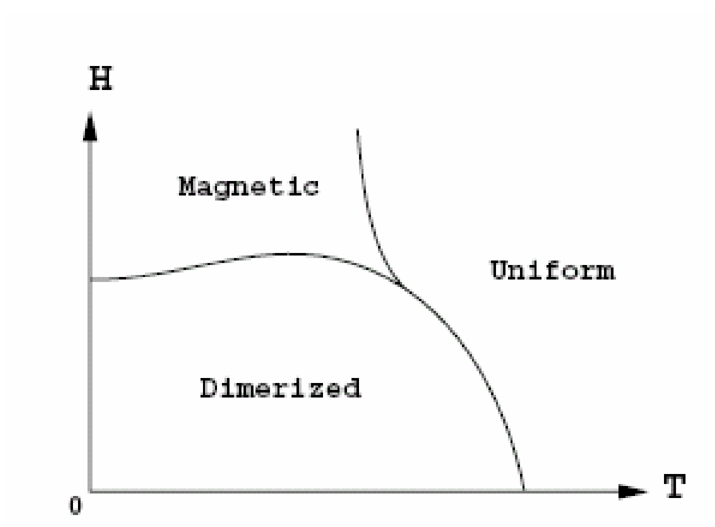


H=0 H>H_C

A regular incommensurate lattice of unpaired spins form in high magnetic fields.

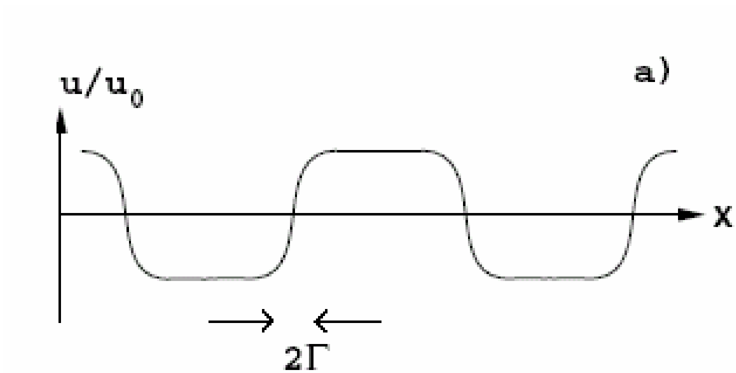
Magnetic field plays a role analogous to chemical potential (band filling) of a regular 1D Peierls system with lattice instability at $2k_F$, where k_F is the field-dependent "Fermi vector"

$$k_F(0) - k_F(\mathrm{H}) = \frac{2g\mu_B \mathrm{H}}{\pi J a}$$

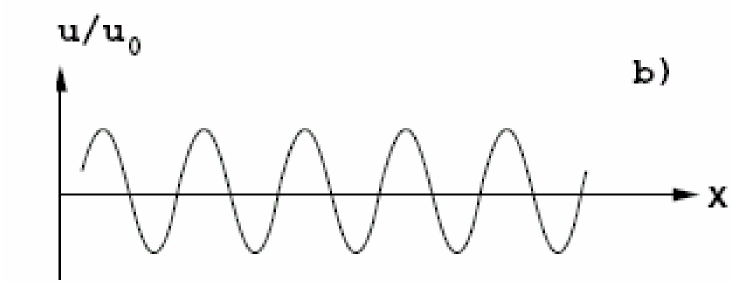


Magnetic Phase Diagram

Effects of a Magnetic Field



Magnetic phase, low field.

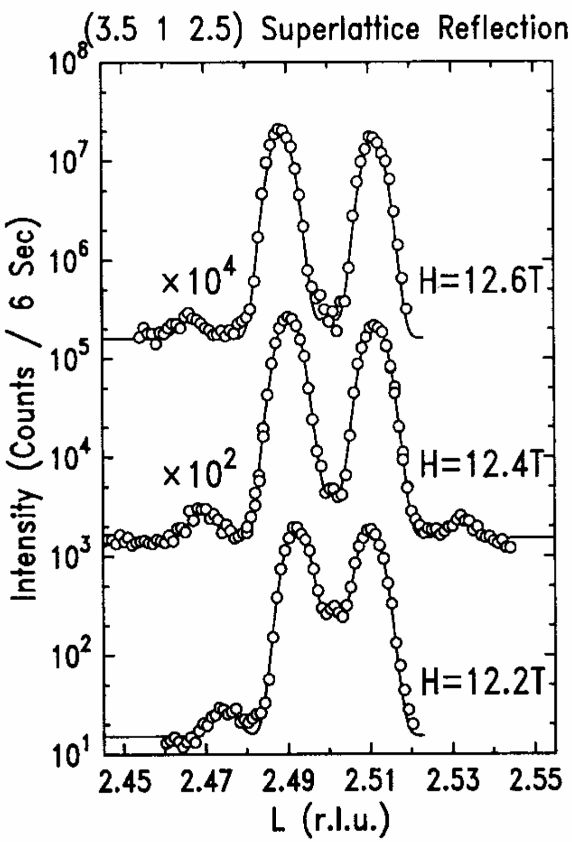


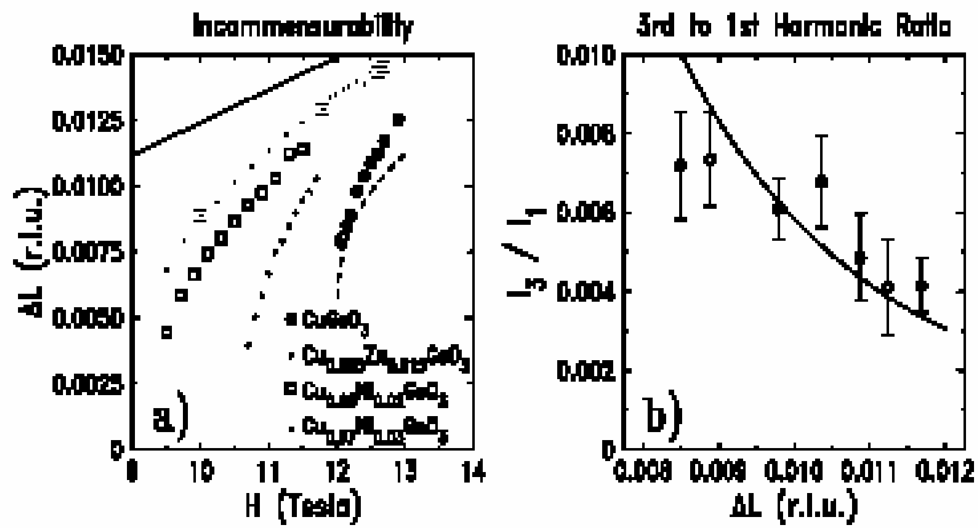
Magnetic phase, very high field.

$$u(x) = u_0(x)\ sn(\frac{x}{\Gamma k}, k)$$

Lattice distortion is not sinusoidal, but is described in terms of the "soliton lattice" – a lattice of regularly spaced domain walls separating the dimerized regions. In very high fields, the lattice modulation approaches the sinusoidal shape.

Effects of a Magnetic Field





The incommensurate soliton lattice is indeed present in high fields. The measured width of the solitonic domain wall, ~11c (larger than theoretical value ~8c). The parameters of the IC state may approach the theoretical values in high fields. Need higher fields.

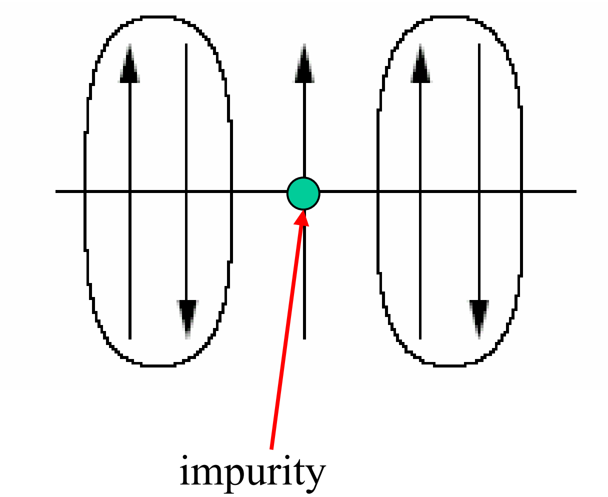
Effects of a Magnetic Field

Questions:

-how is the high-field state approached?

-NMR expts. suggest another anomaly at high fields. Origin?

-effects of impurities?



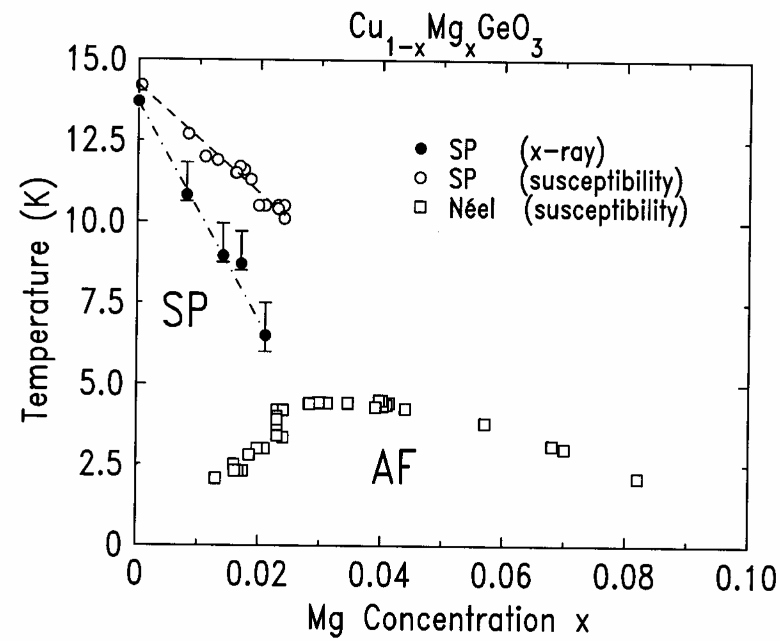
Impurities produce unpaired spins. These spins form an unusual AF state coexisting with the SP spin dimers. How do the impurities affect the field-induced IC solitonic state?

Effects of a Magnetic Field

Disorder Effects in Spin-Peierls Compound $(Cu,Mg)GeO_3$

At zero field, the long-range dimerized state is destroyed at a critical concentration x_c.

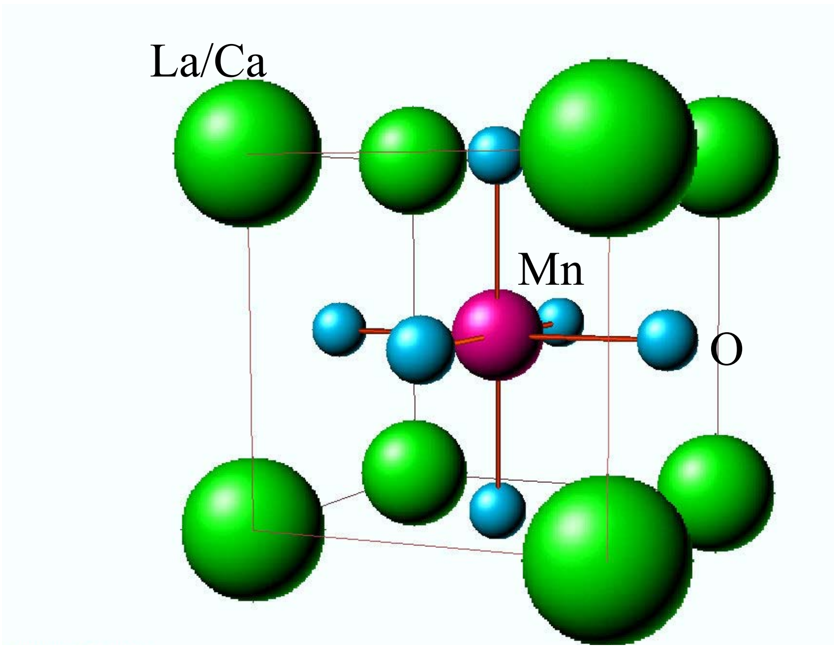
In the incommensurate state, it appears that $x_c=0$, consistent with random-field effects for a system with a continuous XY order parameter, $d<4$.



What happens to the soliton lattice in the disordered samples? Current experiments are limited by low signal.

Research Projects (examples)

2. Magnetoresistive Manganites $A_{1-x}B_xMnO_3$



Electron-electron

Electron-lattice

Superexchange

Long-range elastic strain

e_g: $d_{x^2-y^2}$, $d_{3z^2-r^2}$

t_{2g}: d_{xy}, d_{yz}, d_{xz}

Mn^{3+} $(3d^4)$ Mn^{4+} $(3d^3)$

Colossal Magnetoresistance (CMR)

1. Paramagnetic Insulating

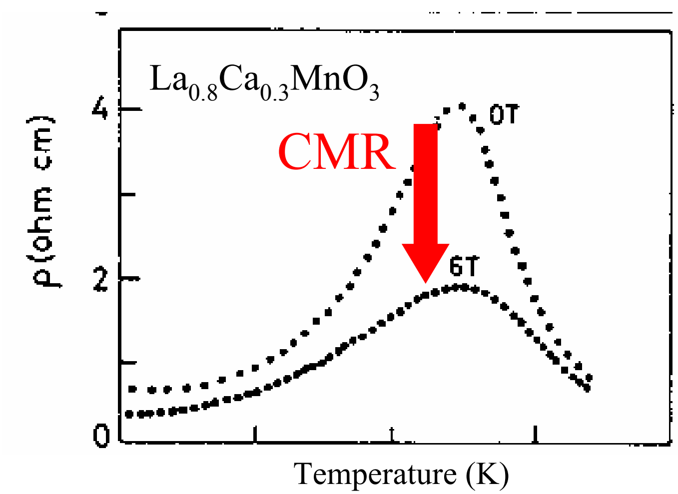
OR

2. Charge-Ordered Insulating

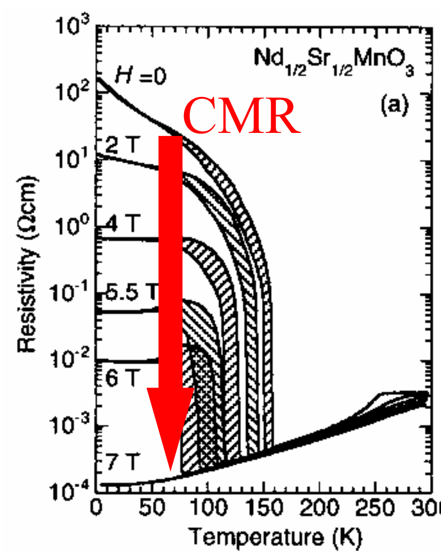
→ Magnetic Field →

Ferromagnetic Metallic State

1. PI to FM



2. CO to FM



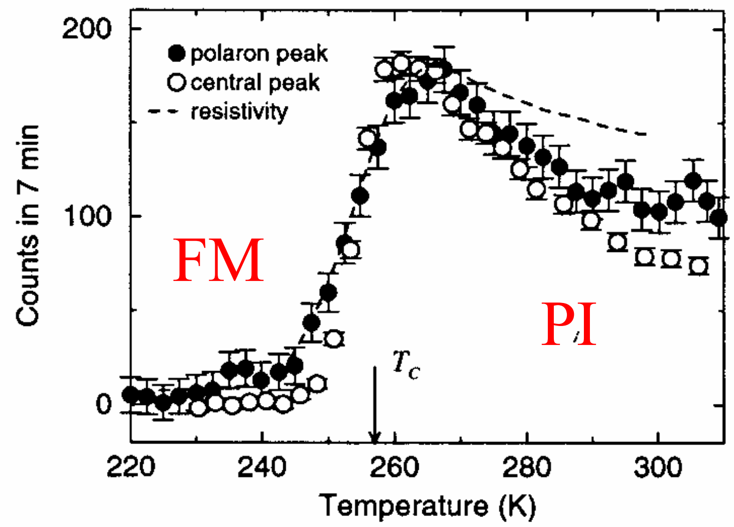
R. Mahendrian, et al., (1996)

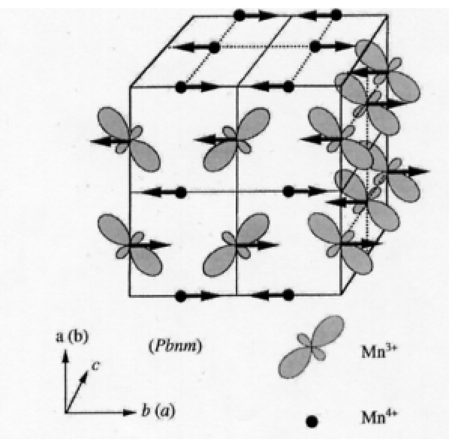
H. Kuwahara, et al. (1995)

Nanoscale Correlations and the CMR

Nanoscale correlated (CO?) regions play an important role.

Broad peaks are observed in neutron scattering experiments at the wavevector characteristic to the CE-type charge/orbital order in the PI phase of $La_{0.7}Ca_{0.3}MnO_3$ and other manganites (Adams, et al.; Dai, et al.; Tokura, et al.; Nelson, et al., 2000).

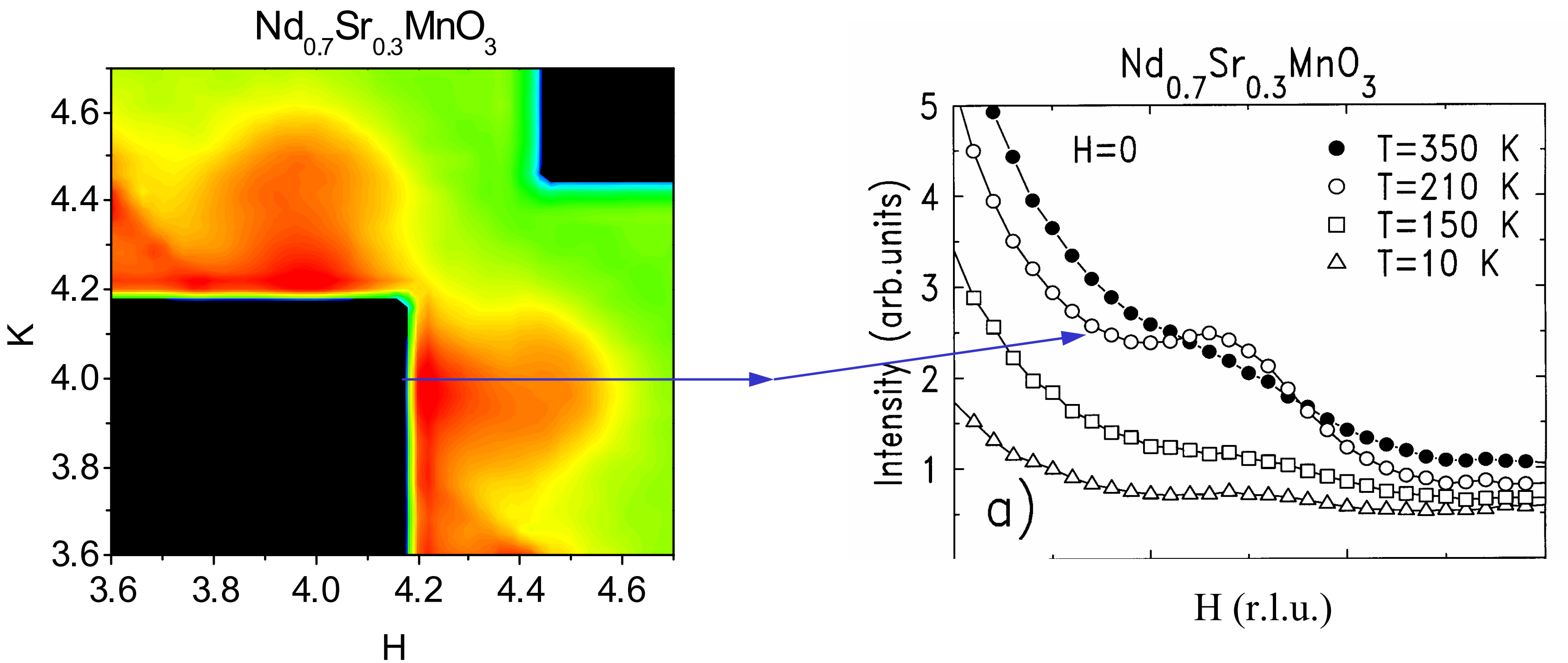




C. P. Adams, et al., (2000)

Nanoscale Correlations and the CMR

x=0.3 samples



Broad peaks at the CE-type position in the PI$^{(*)}$ phase

Nanoscale Correlations and the CMR

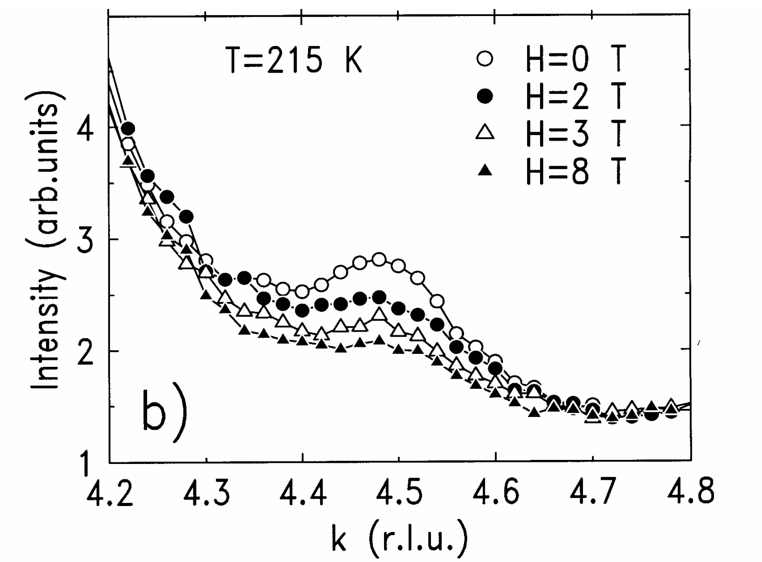
x=0.3 samples

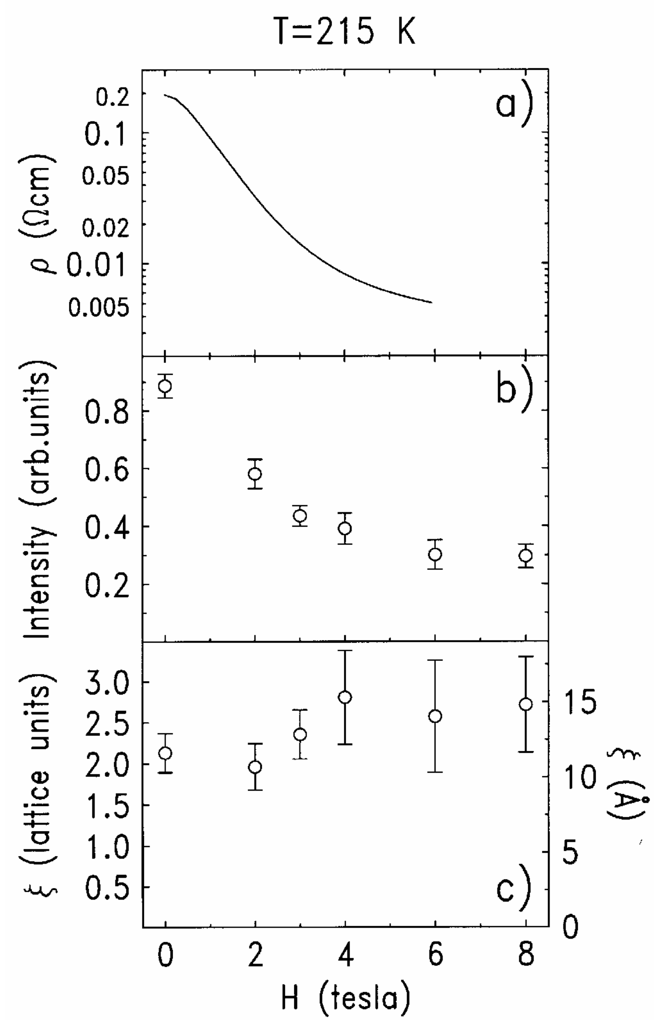
CMR Effect

- correlations suppressed by the field

-inhomogeneous high-field state

- field-independent corr. length





Opposite behavior to the FM clusters observed by SANS

Conclusions

- Nanoscale structural correlations play an important role in the CMR effect. They appear to be at least partially responsible for the large resistivity of the PI state, and for the large value of MR.

-The correlations have common correlation length (10-15 A), their lattice modulation vector has a common linear dependence on the doping level.

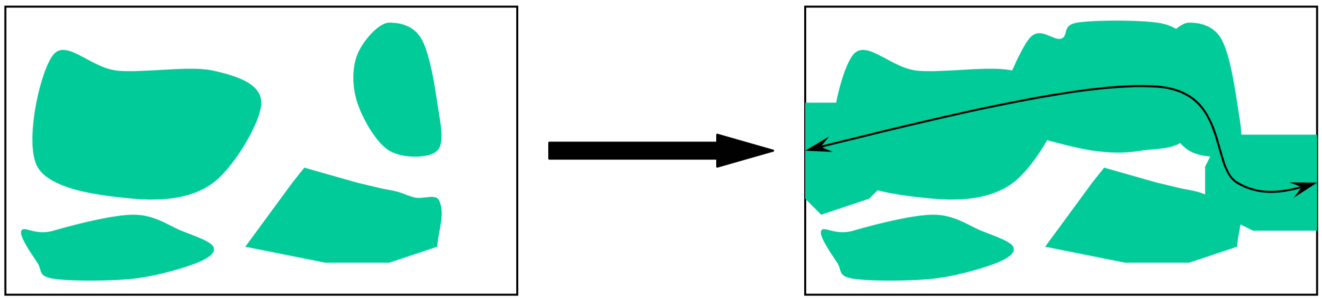
- The structure of the correlated regions is still unknown. The regions may possess charge/orbital order, and might be electron-depleted

- The field-induced state is inhomogeneous.

T. Y. Koo.et al., PRB 64, 220405(R) (2001); C. S. Nelson, et al., PRB 64, 174405 (2001); V. Kiryukhin, et al., PRB 65, 094421 (2002); V. Kiryukhin, et al., PRB 67, 064421 (2003).

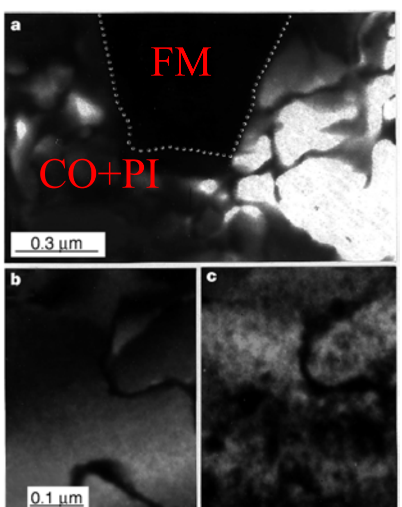
CMR and Inhomogeneous States

1. Phase separation between metallic and insulating phases (~1μm).



Percolative phase transition

(La,Pr,Ca)MnO_3



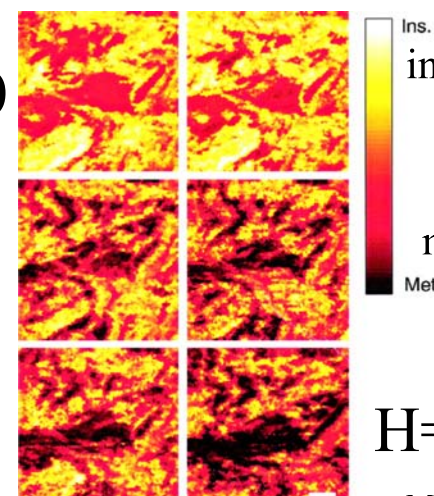
$T<T_c$

$T>T_c$

M. Uehara, et al. (1999)

(La,Ca)MnO_3

H=0



insulator

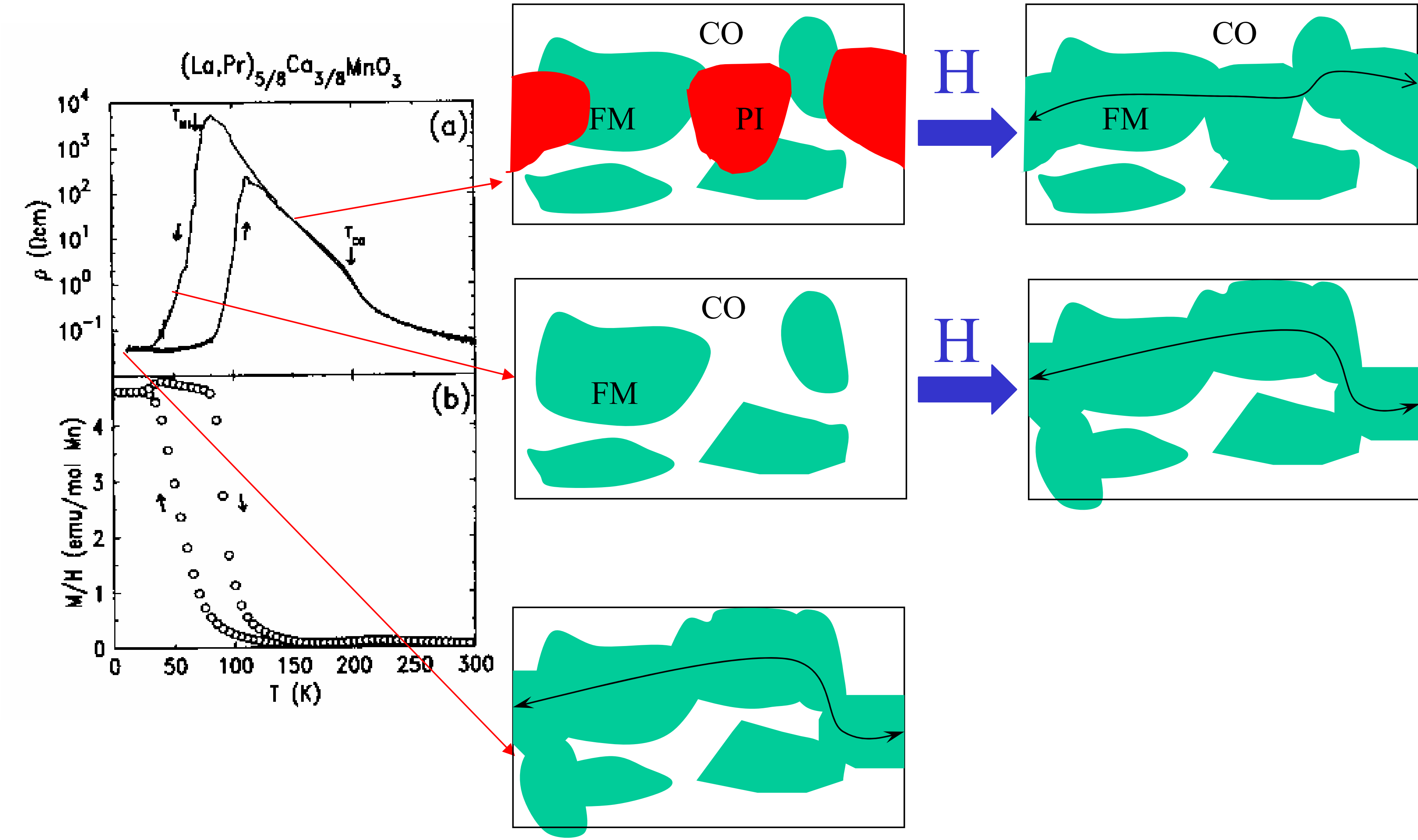
metal

H=9 T

M. Fath, et al. (1999)

CMR and Inhomogeneous States

1. Phase separation between metallic and insulating phases (~1μm).



Charge/Orbital Correlations and the CMR

-What is the actual structure of the correlated regions? Resonant scattering studies of weak diffuse signals.

-What is the nature of the inhomogeneous state in an applied field?

-What is the nature of the field-induced transition in the long-range charge/orbital ordered samples? (In many cases, 3 phases are involved!) Critical scattering, diffuse signals, resonant scattering. Inhomogeneous states.

-Microscopic description of the CMR phenomenon.

Other 3d systems exhibiting charge/orbital order (including short-range): cuprates, nickelates, …

X-ray diffraction studies of strongly correlated systems in an applied magnetic field. Interesting topics.

Why x-rays?

- high resolution
- elemental selectivity (in resonant magnetic scattering, for example)
- can study valence electron density, charge and orbital order
- very small samples
- surfaces and interfaces
- extreme environments possible (high magnetic field, pressure, etc.)
- can study spin and orbital magnetism

-q-dependent electronic excitations, high-energy phonons (inelastic scattering)

- lattice, electronic, magnetic dynamics via coherent scattering

The Sample List

Systems with strong coupling between spin and lattice. Magnetoelectric materials (e.g. $TbMnO_3$.)

Other multiferroic compounds (YMnO3, BiMnO3). General issues of coupling of the dielectric properties and structure with magnetism in these materials.

Magnetic transitions, magnetic phase diagrams. Eg.: rare earth compounds.

Physics of frustrated magnets (spinels, pyrochlores, etc). Critical properties, static correlations.

Structural phase transitions in magnetic fields. Studies of critical properties, effects of phase competition, etc. Eg.: Bicritical properties of CoS(2-x)Se(x).

Physics of charge and orbitally ordered states (manganites, ruthenates, vanadites, cobaltites,…), and field-induced transitions in these compounds. Studies of mixed-valent compounds.

Jahn-Teller systems (manganites).

Physics of correlated materials with strong phase competition and intrinsic nanoscale inhomogeneous states. Materials with large responses to an applied field. (Manganites, cuprates (?)).

Related topic. Structural properties of magnetic semiconductors. Phase separation in these materials.

Physics of quantum critical point. Determination of structural and magnetis states in an applied field. Eg.: URu2Si2, ruthenate compounds.

Related topic: magnetism in heavy-fermion materials and in exotic superconductors. Field induced structural and magnetic transitions. Eg. UPt3, CeRu2Si2.

Physics of high-Tc superconductivity. Nature of the normal state at high fields (magnetism). Phonons.

Fundamental physics of Mott transition. Excitation spectrum of mott insulators in the vicinity of the first-order Mott transition in an applied field. Oxide and sulfur based compounds, BEDT-TTF-based organics, etc.

One-dimensional magnetism. Spin-Peierls compounds (CuGeO3 well above Hc), field-induced density waves in the TMTSF-based compounds, field-induced incommensurate states in Cu(Hp)Cl. Studies of physics of commensurate-incommensurate transitions.

Currently available facilities for x-ray/neutron diffraction in a magnetic field.

a) X-ray scattering. 13 Tesla steady state superconducting magnets at the NSLS, APS, SSRL. A very recent addition: a 15 Tesla magnet at Spring-8. Full q-range, temperature range 1.5-300 K (lower T at the Spring-8 magnet).

b) Neutron scattering. 15 (17) Tesla DC magnet at Hahn-Meitner Institute, Germany. Various 12 Tesla magnets at other facilities. A 17 Tesla pulsed magnet (*ms* pulses) at Kobe/KENS, Japan.

Proposed facilities. A steady state 30-40 Tesla non-superconducting magnet at HMI (status uncertain). A 30 Tesla pulsed field (3 *ms*, 2 *Hz*) magnet, developed by NHMFL/LANSCE. Neutron diffraction.

Diffraction in high magnetic field at the NSLS.

<u>The Original Setup at X22B</u>

-A 13 Tesla vertical field superconducting magnet (one of only a few in the world)

-Bending magnet beamline, single Ge monochromator, E=6-10 keV

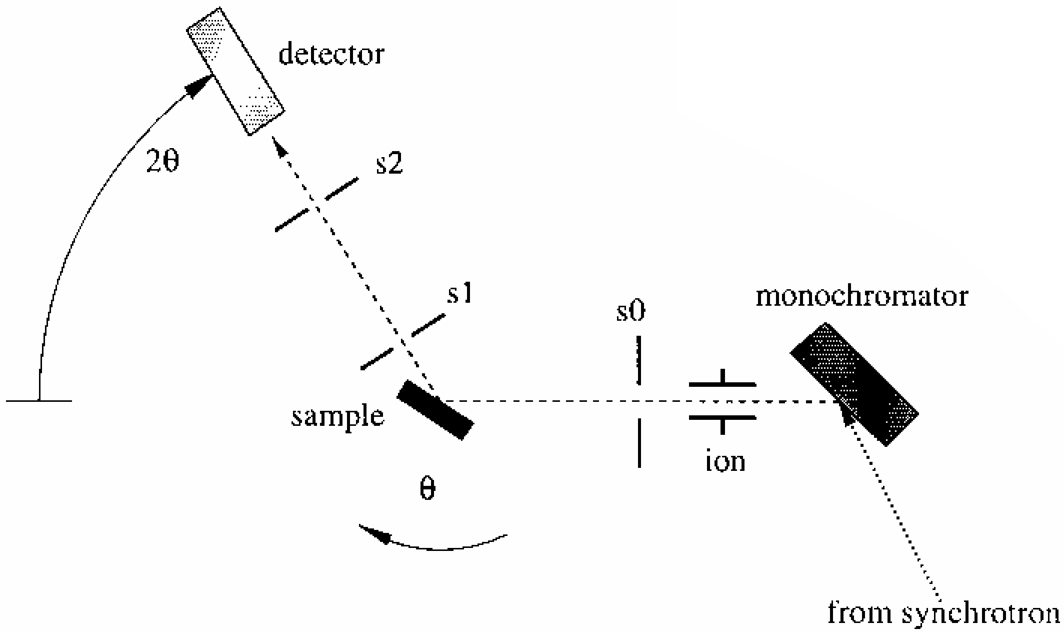
-Horizontal scattering geometry

-Two-axes spectrometer, no tilt stages

-The instrument is not permanently located in the experimental hutch and needs to be moved in before every experiment. Helium transfers are very difficult. Accessible q-range is limited by the small hutch dimensions for most of the x-ray energies.

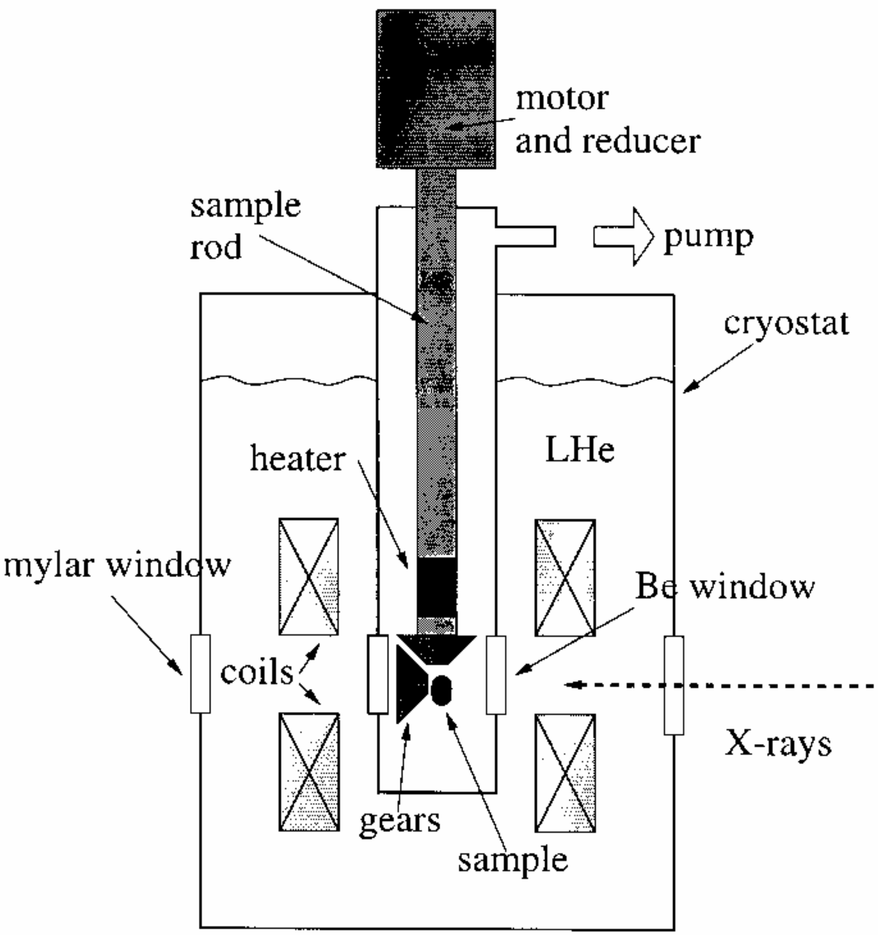
-No dedicated experimental support, which is urgently needed to maintain the magnet operational. No general user support.

X-ray diffraction in a high magnetic field at x22b



- bending magnet beamline
- horizontal scattering setup
- Ge (111) monochromator
- ~6-10 keV energy range
- ~5 x 10^{10} photon flux
- two-circle diffractometer

X-ray diffraction in a high magnetic field at x22b



-13 Tesla superconducting magnet (owned by MPI-Stuttgart)

- vertical magnetic field (perpendicular to the scattering plane)

- 1.6 K – 300 K sample temperature

- possible to fill the sample space with liquid helium

- in-citu sample rotator

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X-ray diffraction in a high magnetic field at x22b

INVSETIGATED SYSTEMS

-spin-Peierls materials ($CuGeO_3$, TTF-(Cu)BDT, $(TMTTF)_2X$)

- CDW systems ($NbSe_3$, $NbSe_2$)

- magnetism in rare earths (Ho, Dy, Er, etc.): magnetic resonant x-ray scattering at L-edges

- FISDW $(TMTSF)_2X$

- CMR manganites: charge and orbital order, inhomogeneous states, nanoscale correlations, x-ray-induced structural/magnetic transitions

The upgraded facility at x21

A new permanent *user* facility for x-ray scattering in high magnetic fields

High-flux wiggler beamline. Flexible setup

-Si (111) double-crystal monochromator: E=3-24 keV, ΔE/E=0.05%, flux more than 10^{12} s^{-1}.

- High-energy resolution monochromator: E=5.5-9.2 keV, ΔE=0.2 eV, flux ~5 x 10^{10} s^{-1}.

- Multilayer mono: E=6-13 keV, ΔE~100 eV, flux ~10^{14} s^{-1}.

Better spectrometer, instrument and user support.

The upgraded facility at x21

Important advantages over the existing facility (a partial list):

-High flux. Flux-limited systems: disordered systems, small crystals, non-resonant magnetic scattering, diffuse scattering.

-Energy tunability and energy resolution. Resonant scattering studies of charge and orbital order, resonant magnetic scattering. 3d and 5d systems, rare earths, actinides.

-Better spectrometer – increased range of q available.

-Instrument support. User support – general users.

What experimental capabilities are needed?

Questions:

- field: continuous, pulsed, what range?
- energy range (resonant studies, penetration depths)
- momentum an energy resolution (resonant expts, inelastic)
- flux (weak signals, diffuse scattering, polarization analysis, inelastic)
- . . .

Several thoughts on this subject.

For a core set of instruments, a steady state, high-field magnet is needed. Desired features: temperature range of at least 0.3-300 K (or an even larger range); large bore to accommodate sample environment devices, such as high-pressure cells; windows suitable for small-angle scattering and inelastic experiments (low background), and for lower x-ray energies (low adsorption). Such an instrument will permit a large set of experiments, including single-crystal and powder diffraction, resonant scattering, magnetic diffraction, inelastic x-ray scattering, small-angle scattering measurements, etc. It is expected that with a suitable technical support, it will become a highly utilized and versatile user facility.

The steady state magnetic field produced by superconducting magnets appears to be limited by ~20 Tesla. Higher fields (in an acceptably compact instrument) can be achieved in a pulsed-field magnet. Pulsed magnets utilize less than 1% of the steady-state x-ray source intensity. In addition, while pulsed magnets for neutron scattering do exist (and a 30 Tesla magnet is under development now), no working design for x-ray scattering has been demonstrated, to my knowledge. (I would appreciate any info on that, if it is available). Despite these drawbacks, a pulsed magnet instrument appears to be the best solution for high magnetic field x-ray scattering experiments at this stage. Usefulness of such a facility will be limited by the low effective x-ray flux, and studies of systems exhibiting hysteresis, ageing effects, and other time-dependent properties will be severely restricted.

In conclusion,

- x-ray scattering in an applied magnetic field is a very useful experimental technique. A large number of scientific opportunities and interesting projects do exist.

- a steady-state high-field magnet and a dedicated beamline should, in my opinion, be included in a core set of instruments for the NSLS II.

- the parameters of this beamline should be a subject of discussion involving a large number of scientists.

- open question: is there a compelling justification for a pulsed field facility at a synchrotron?

- last but not least, it is important to identify other interesting scientific projects involving x-ray scattering and magnetic fields.