

Time-of-flight Spectrometer for Diffuse Neutron Scattering (DNS)



Instrument Parameters

Beam tube:	NL-1, straight neutron guide, 10 cm (height) $\times$ 7 cm (width)
Monochromator:	double focusing graphite, 6 crystals, each $5 \times 4 \times 0.2 \text{ cm}^3$
Wavelength:	$3.3 \text{ \AA} < \lambda < 5.5 \text{ \AA}$
Energy:	$7.4 \text{ meV} > E_{\text{initial}} > 2.7 \text{ meV}$
Energy resolution:	$0.9 \text{ meV} > \Delta E > 0.15 \text{ meV}$ (FWHM, elastic)
Momentum transfer:	$< 3.5 \text{ \AA}^{-1}$ (elastic)
Vertical:	$< 0.5 \text{ \AA}^{-1}$ (elastic)
Polarizer:	focusing supermirror FeCoV/Ti:N
Transmission:	30 % to 50 % for correct spin
Sample position - Neutron flux: - Beam size: - Special sample - Environments:	$(10^7 \dots 3 \times 10^6) \text{ n/cm}^2\text{s}$ max. 6 cm (height) $\times$ 2 cm (width) split-coil magnet 5 tesla with dedicated cryostat, $T > 2 \text{ K}$ furnace $T < 1600 \text{ }^\circ\text{C}$, sample rotation inside
Detectors:	52(+12) ^3He -detectors (4 bar), solid angle: 0.3 sr
Angular range:	$0^\circ < 2\Theta < 135^\circ$
Vertical:	$2\Theta < 15^\circ$
Distance to sample:	80 cm
Options for analysis - Time-of-flight: - Bragg: - Polarization:	disk chopper, duty cycle 0.05 104 graphite analyzers for $E_{\text{final}} = 7.4 \text{ meV}$ 12 detector unit with polarisation analysis; focusing, supermirrors FeCoV/Ti:N

Instrument Responsible

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Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-02-009		
Experiment title:	Investigation of Magnetic and Structural Correlations in the diluted semiconductor $\text{Ga}_{1-x}\text{Mn}_x\text{As}$ ($x=0.05$)		
Dates of experiment:	Dec. 2004	Date of report:	May 2005
Experimental team: Names	Addresses		
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Local Contact:	W. Schweika		

Experimental report text body

The magnetism of MnAs shows evidence for a strong coupling of magnetic and lattice degrees of freedom. The transition from a ferromagnetic to a paramagnetic phase proceeds via a 1st order phase transition in which the crystallographic structure changes discontinuously from a hexagonal space group at low temperatures to an orthorhombic one at higher temperatures. The change in the size of the magnetic moment is significant. Just below the transition the ordered magnetic moment is ~70% of the ground state moment. After the crystallographic transformation the substance is paramagnetic with strong ferromagnetic short range order correlations.

For diluted compounds, in which MnAs is substituted into a matrix of GaAs, also shows magnetism and ferromagnetic ordering. The amount of MnAs which can be dissolved in the matrix is limited to a few percent. The amount of MnAs can be increased for thin film samples, while bulk material is limited to approximately 5% maximum. Despite the high degree of dilution in a non-magnetic matrix the transition temperatures are surprisingly high, exceeding 100K for optimally prepared thin film samples.

In order to investigate the formation of a macroscopic ferromagnetic state for a diluted MnAs sample a neutron scattering experiment has been carried out in order to determine the nuclear and magnetic scattering as a function of temperature.

The scattering at high temperatures (300K) and at low temperatures (10K) are shown in figures 1 and 2.

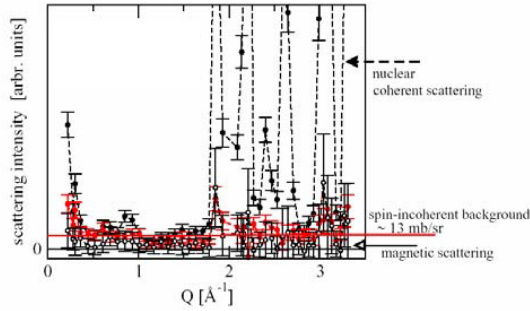


Figure 1: scattering of $\text{Mn}_x\text{Ga}_{1-x}\text{As}$ at $T=300\text{K}$.

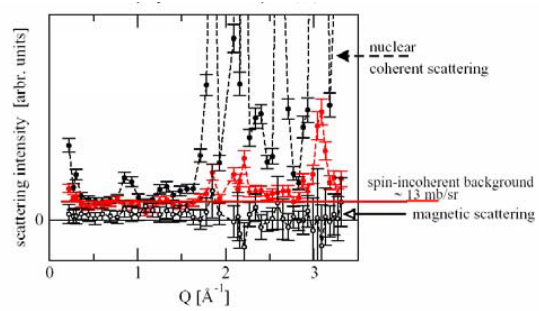


Figure 2: Scattering of $\text{Mn}_x\text{Ga}_{1-x}\text{As}$ at 10K

A particular problem with the investigation of these samples is phase separation and the segregation of a ferromagnetic MnAs phase. This results in ferromagnetic inclusions at low temperatures which prevent spin polarized neutrons from being used to probe magnetic correlations. As figures 1 and 2 demonstrate spin polarized neutrons can be used to probe diluted $\text{Mn}_x\text{Ga}_{1-x}\text{As}$ samples, if sufficient care is exercised in the preparation of these samples. However, due to the low concentration of magnetic scatters the measurement time for each sample has to be much longer.

The experiment has looked at several samples differing in the concentration of Mn. For samples with a high concentration the problem of depolarization of the neutron beam arises. For samples with a low concentration and with the appropriate heat treatment, suitable samples for bulk measurements are available for neutron scattering experiments. A substantial proportion of the experimental time was used to establish and verify the optimal experimental conditions. The resulting magnetic correlations, however, have been obtained only with a large error bar such that on the basis of the initial measurements no definite statements can be made as to the temperature dependence of magnetic correlations.

In conclusion the experiment confirms the feasibility of measuring magnetic correlations in diluted ferromagnets such as $\text{Mn}_x\text{Ga}_{1-x}\text{As}$. However, longer measurements are needed in order to establish details of the magnetic correlation functions and their temperature dependence.



Experimental Report of Neutron Scattering Experiments at the FRJ-2 Reactor

Proposal number:	DNS-04-011		
Experiment title:	Low-frequency dynamics of spin and orbital polarons in ferromagnetic insulating manganites		
Dates of experiment:	21-04-06 to 02-05-06	Date of report:	06-06-06
Experimental team: Names	Addresses		
Dr. Y. Su	Institut für Festkörperforschung, Forschungszentrum Jülich		
Local Contact:	Dr. W. Schweika		

Experimental report text body

“1/8”-doped $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x \approx 1/8$) is among the most fascinating in a large class of highly correlated transition-metal oxides, where the interplay between charge, spin, orbital and lattice degrees of freedom plays a key role in determining their highly unusual properties. When a small amount of holes are introduced into A-type anti-ferromagnetic LaMnO_3 via chemical substitutions, a ferromagnetic insulator (FMI) emerges at low temperatures. While the experimental evidences strongly suggest the existence of complex charge/orbital ordering (CO/OO) in the FMI regime [1], the exact nature of CO/OO is yet to be fully established. The fingerprints of possible CO/OO can also be reflected from the nature of magnetic correlations. For example, a change of the anisotropy of exchange coupling (J_{ij}) is expected at the OO transition [2]. In addition, the doping induced holes often have a strong tendency to be segregated into hole-rich or hole poor regions to minimize Coulomb interaction and magnetic frustration, so that the corresponding exchange coupling may also be dramatically altered. One famous example is the occurrence of intriguing spin fluctuations due to the stripe order in high- T_C superconducting copper oxides [3]. The magnetic exchange constants can usually be obtained via measuring spin-waves excitations in a magnetically ordered phase below the Curie temperature (T_C). In the paramagnetic phase above T_C , it is also possible to extract exchange constants from a quantitative fitting to a diffuse magnetic scattering pattern [4].

Comprehensive investigations of magnetic correlations of this peculiar compound have been undertaken on a large single crystal at time-of-flight spectrometer DNS. Both diffuse magnetic scattering and spin-wave excitation were obtained with the neutron wavelength of $4.8 \text{ \AA}$ over a large temperature range. As shown in Fig.1, in the ferromagnetic metallic regime ($155 \text{ K} < T < 190 \text{ K}$), the diffuse magnetic scattering is found to show a prominent *rod*-like feature extended along the $[100]_C$ direction in the reciprocal space of a pseudo-cubic setting. Due to the presence of severe twinning in this perovskite compound, e.g. $(110)/(001)$ twinning in the

orthorhombic setting, the observation of this *rod-like* diffuse scattering strongly suggests the presence of a significant anisotropy of magnetic correlations in this compound, i.e., strong exchange coupling in the orthorhombic *a-b* plane (J_{ab}) and weak inter-plane coupling (J_c). In addition, the low-energy spin-wave excitation was also measured. In contrast to the results obtained in several recent investigations via triple-axis spectrometer [5-6], no splitting in the spin-wave branch can be observed up to 10 meV, as shown in Fig.2. Complete data treatment is in progress.

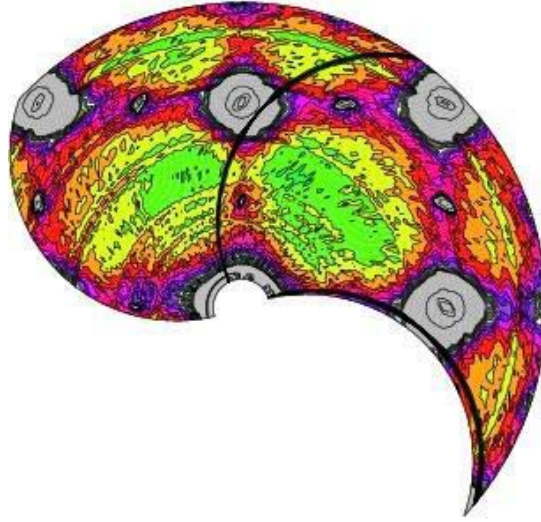


Fig.1 Diffuse magnetic scattering measured at 170 K. (Data shown is the integration over the time channels of all elastic lines.)

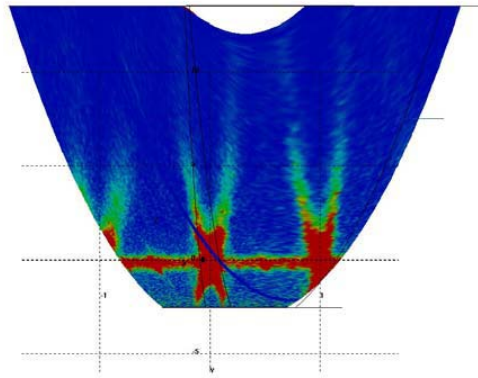


Fig.2 Spin-wave braches measured at 120 K. The horizontal axis is Q , the vertical one is E (mev).

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- [2] Y. Endoh, *et al.*, Phys. Rev. Lett. **94**, 017206 (2005)
- [3] J.M. Tranquada, *et al.*, Nature, **429**, 534 (2004)
- [4] T. Chatterji, *et al.*, Phys. Rev. B **65**, 134440 (2002)
- [5] M. Hennion, *et al.*, Phys. Rev. B **73**, 104453 (2006)
- [6] Y. Su, *et al.*, unpublished



Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-04-065		
Experiment title:	P re aus o rf s tahf iopl tt acf i rots e nhutR rDRl o ital f octhf orf r El xms b dnl f e rEl y41 a24ms b d		
Dates of experiment:	. arch 60 th -20 th 2005	Date of report:	. arch 86 st 200A
Experimental team: Names	d l lresses		
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, he liscoKerl of exotic superconluctiKitl in Sr_2Ruy_4 “: aenoA6””4] anl its structural similaritl to é a₂Buy₄Aa parent compoundl for the high-temperature superconluctorsAhas generate1 much interest in the Ru1lleslen-Popper vRP[series of ruthenium oxilesAvSr]Ba[_{ll u}Ru_y _{sll u} “NakatsuGA6””””A2000A2006WFrie1tA2006WGalknA2006Aé eeA2002]. , he similarities betCeen Sr_2Ruy_4 anl high-temperature superconluctors are best emphasize1 bl consileration of the famill of compoundlsABa₂₋₄Sr₇Ruy₄ACHose crlstal structure is shoCn in Figure 6. , he experimentalll obserKel behaKior for these compoundls as a function of lopingAshoCn in Figure 2Ahas remarkable similarities Cith the first obserKel high-temperature superconluctorA é a₂₋₄Sr₇Buy₄. . oth sets of compoundls haKe an insulating antiferromagnetic vLF. [grounl state vLFv] in Fig. 2[Aa structural phase transition separating high temperature tetragonal vH, , [anl loC-temperature orthorhombic vé, y [phases vH, , -é, y in Fig. 2[Aa paramagnetic metallic phase at larger Kalues of τ vPw [-tet in fig. 2[Aanl a superconlucting phase vSB in fig. 2[. In al lition to proKiling a neC slstem Chere the eKolution from antiferromagnetism to superconluctiKitl can be explore1Athe phase liagram of $\text{Ba}_{2-4}\text{Sr}_7\text{Ruy}_4$ contains rich anl exotic behaKior attributel to structural anl magnetic instabilities.

, he first obCctiKe of this proCct is to obtain the magnitule of the magnetic moments in Ba_2Ruy_4 anl $\text{Ba}_{6.5}\text{Sr}_{0.5}\text{Ruy}_4$ anlA furtherAto obtain real-space spin-spin correlation function from the Fourier-transform of the magnetic structure factor. , he seconl obCctiKe is to unlerstanl the tetragonal to orthorhombic structural phase transition associatel to the rotation of the Ruy_A octahelra aroundl the z-axis in $\text{Ba}_{6.5}\text{Sr}_{0.5}\text{Ruy}_4$ Athrough scattering on a single crlstal. , he origin of this lattice leformation is not unlerstoodAanl a liffuse scattering stull is expectel to liell insight into the mechanism of this phase transition.

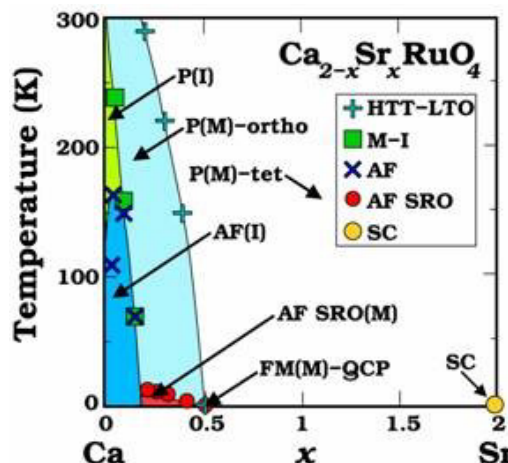
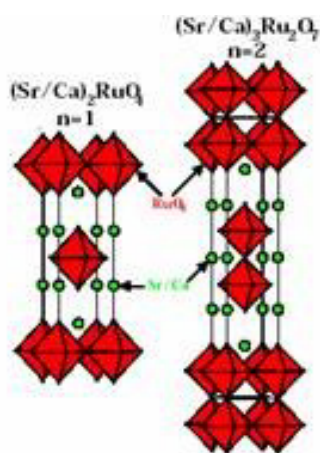


Figure 4: Schematic crystal structure of the Y6 Ba_{2-x}Sr_xRu_{1-y}Ru_yO₄ system.

Figure 6: Phase diagram of the Y6 Ba_{2-x}Sr_xRu_{1-y}Ru_yO₄ system.

The magnitude of the magnetic scattering in Ba_{6.5}Sr_{0.5}RuO₄ has proven too small to obtain any information on the spin correlation in that material. In Ba₂RuO₄ the only significant magnetic scattering observed has been observed at $Q = 0.688 \text{ \AA}^{-1}$ and at $Y = 600 \text{ q}$. As seen in Figure 8, this peak is absent at 200 q . During this series of measurements it has appeared that the magnitude of the spin incoherent scattering in both compounds is extremely different, suggesting that the amount of humidity that these compounds trap is much larger than previously believed.

The second part of the measurements will be the expected result because the quality of the single crystal is not sufficient so that we could not collect a usable inelastic signal from the phonon scattering at low temperature.

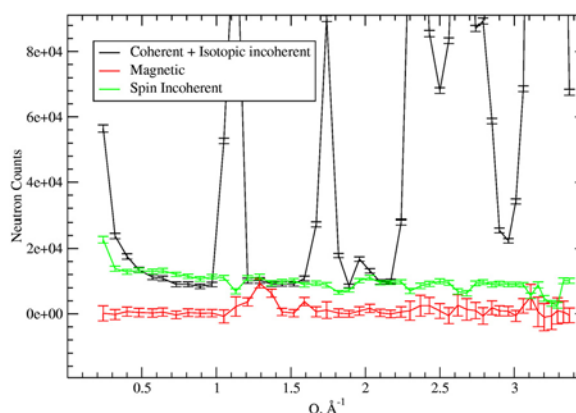
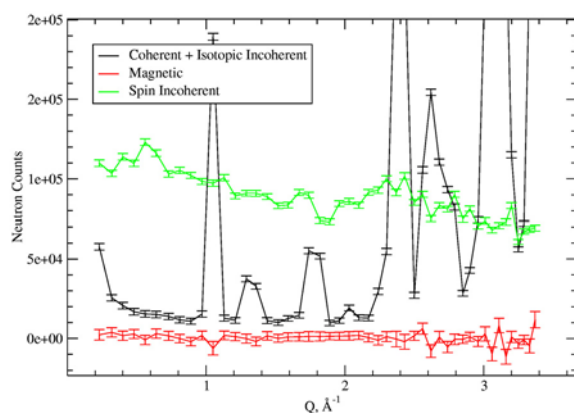


Figure 3: Neutron scattering data for Ba_{6.5}Sr_{0.5}RuO₄ at 600 q (left) and Ba₂RuO₄ at 600 q (right).

R u a f p o 6

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Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-04-06.		
Experiment title:	Preparation of a single crystal of $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ for neutron scattering experiments		
Dates of experiment:	8 April to 12 June 2005	Date of report:	8 March 2006.
Experimental team: Names	Addresses		
Hermann Rapp, Jean-François Granado, Egami Kakeshi, Stercel Ferenc	Université de Liège, Dept. de Physique, s.d. 4000 Liège, Belgium Université de Liège, Dept. de Physique, s.d. 4000 Liège, Belgium University of Tennessee, Dept. of Materials Science and Engineering, Knoxville, TN, USA University of Tennessee, Dept. of Materials Science and Engineering, Knoxville, TN, USA		
Local Contact:	Dr. T. Schenck		

Abstract

In the cuprate superconductor system $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ an antiferromagnetic long-range order disappears at $x \approx 0.02$, and a superconducting phase appears above $x \approx 0.01$. At low temperatures the region in-between, $0.02 > x > 0.01$, is called the spin-glass phase. However, the real nature of this phase is not clear. Furthermore spin-glass-like slow magnetic spin correlation is suspected to persist up to high temperatures and high doping concentrations over the so-called pseudo-gap region, but again its real nature is poorly understood. The most important question is how the spins confine the doped charges. We have made the system insulating, or poorly conducting. It is generally believed that the charges are confined in the stripe state in this concentration range $x \approx 0.02$, just as in the $\text{NdO}_{1-x}\text{F}_x$ samples $x \approx 0.02$. However, the evidence is only indirect and confusing. The purpose of this measurement is to determine the true spin configuration in this composition range and determine how spins are confining charges.

The objective of this project is to determine the spin correlation in $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ for $0.02 > x > 0.01$ as a function of temperature and doping, x , and how the spin correlation deteriorates with increasing temperature. This measurement is expected to give information on the spin-glass phase that is suspected to exist near the superconducting phase in the $\text{La}_{1-x}\text{Sr}_x\text{CuO}_4$ phase diagram.

In the so-called pseudo-gap phase under investigation it has been proposed that there exists a mixture of nano-scale AF clusters and possible paired holes, but this conjecture is based upon the macroscopic measurements of spin susceptibility and the NMR spin-lattice relaxation time T_1 vs. frequency. Here we intend to conduct direct measurements—

Diffuse magnetic scattering has been observed in $\text{La}_{6-14}\text{Sr}_{0.0}\text{Ba}_{4-x}$, indicating the presence of a spin-glass phase in this compound at 2 K and 20 K , see Figure 6. Given the small magnitude of the magnetic scattering with respect to both the nuclear isotopic incoherent and nuclear spin incoherent scattering, the analysis of the data requires extremely good determination of all ancillary parameters, such as background, flipping-ratios, and absolute scattering. In order to assure both reproducibility and statistical reliability of the data, we have preferred to focus on only three samples, namely $\text{La}_{2-x}\text{Sr}_x\text{Ba}_{4-x}$ with $x \sim 0, 0.0$, and 0.6 . The first series of measurements has been conducted at $L\text{-L } \text{\AA}$, in order to investigate the largest range of scattering vectors possible. However, the presence of many strong Debye-Scherrer rings at larger Q spoils any possibility to obtain a good separation of the magnetic and incoherent scattering. Further these Debye-Scherrer rings are strong multiple scattering that taints the data. Hence, for the most interesting sample, $x \sim 0.0$, the results have been reproduced at a larger Q -length of $4\text{ \AA}^{-1}$. This measurement has the advantage of both enhancing the resolution and of reducing the multiple scattering and has shown the presence of the spin-glass phase—

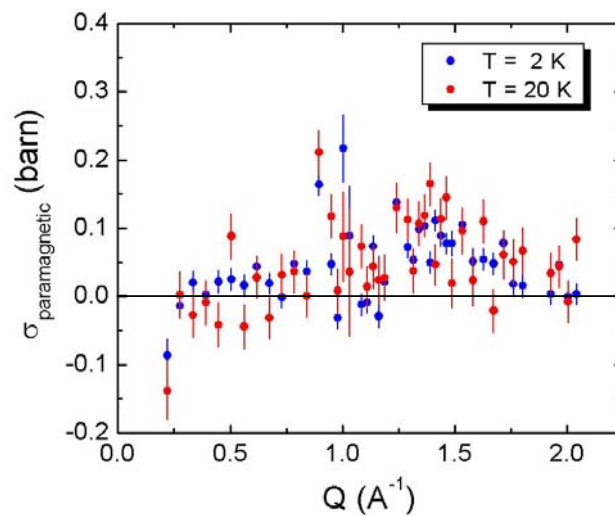


Figure 6 Paramagnetic scattering in $\text{La}_{6-14}\text{Sr}_{0.0}\text{Ba}_{4-x}$. The noise just below 6×10^{-6} is caused by a Debye-Scherrer ring—

References

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Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-04-017		
Experiment title:	A diffuse neutron scattering study of phase transitions in $\text{Na}_{0.75}\text{CoO}_2$		
Dates of experiment:	February 28 th - March 9 th 2005	Date of report:	March 31 st 2006
Experimental team: Names	Addresses		
Hermann Raphaël, Grandjean Fernande, Keppens Veerle, Mandrus David	Université de Liège – Dept. de Physique – B5 – B4000 Liège – Belgium Université de Liège – Dept. de Physique – B5 – B4000 Liège – Belgium University of Tennessee, Dept Material Science and Engineering, Knoxville, TN, USA Oak Ridge National Laboratory, Oak Ridge, TN, USA		
Local Contact:	Dr. W. Schweika		

Experimental report text body

The layered transition metal oxides, Na_xCoO_2 , have been of considerable interest to the scientific community for the past few years. This family of materials has been known for some time¹ but the report by Terasaki et al.² concerning possible thermoelectric applications has rekindled interest in these compounds³ It is very unusual for an oxide to have a relatively high thermopower yet a low metallic resistivity⁴. This result alone suggests interesting electronic correlations. The subsequent discovery of superconductivity at 5 K by Takada et al.⁵ in a hydrated version of this material ($\text{Na}_{0.35}\text{CoO}_2 \cdot 1.3 \text{ H}_2\text{O}$) caused even greater excitement. Although the superconducting transition temperature is too low for applications, the structural and chemical similarities between Na_xCoO_2 compounds and cuprate superconductors provide hope of additional insight into the physics of superconductivity in layered transition metal oxides. The basic structure of Na_xCoO_2 ($0.3 < x < 0.9$) consists of planes of edge-sharing CoO_6 octahedra alternating with layers of Na. There are two crystallographically distinct Na sites, both of which are only partially occupied. In Na_xCoO_2 , the Co ions in each plane form a triangular lattice rather than the square lattice found in the cuprates. A $S=1/2$ triangular lattice is of great interest to theorists because of the inherent geometrical frustration of antiferromagnetic order and since Anderson's original RVB model for superconductivity⁶ was calculated for a triangular lattice. This experiment is focussed on single crystals and powder with $x \approx 0.75$. Crystals with this composition undergo an unusual magnetic spin-density-wave transition near 22 K, an order-disorder transition near 340 K and several subtle transitions at intermediate and lower temperatures that have recently been mapped by us using resonant ultrasound spectroscopy.

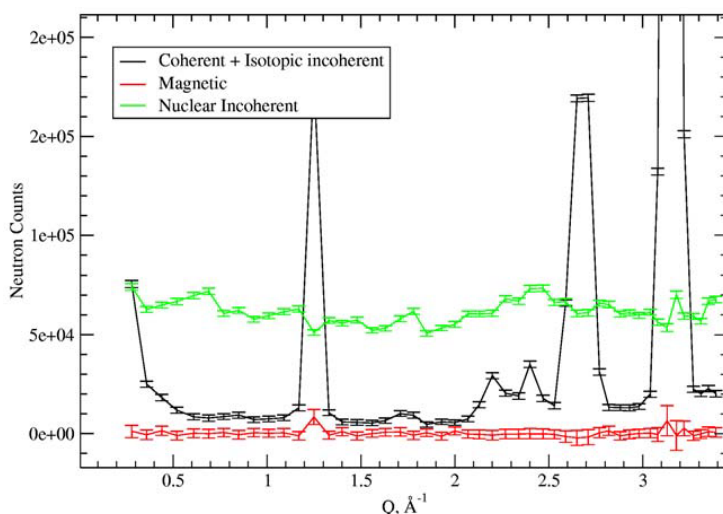


Fig. 1: Scattering by $\text{Na}_{0.75}\text{CoO}_2$ powder at 1.8 K.

The first goal of this project is to determine the real-space spin-spin correlation function in the novel complex metal oxide $\text{Na}_{0.75}\text{CoO}_2$ via a determination of the magnetic diffuse scattering across the antiferromagnetic phase transition, using polarization analysis. The second goal is to determine the spin excitation spectrum, and possible the magnetic exchange constants via an energy spectrum using time-of-flight analysis. The polarization analysis of the neutron scattering by $\text{Na}_{0.75}\text{CoO}_2$ powder at low temperature has not revealed any significant magnetic contribution, see Fig. 1. This permits one to set a limit on the maximum magnitude of the Co moments, but doesn't give any information on the magnetic correlations in the material. Given this result, it was decided that an investigation of the excitation spectrum was not feasible under these condition.

Instead it was preferred to measure the scattering by a $\text{Na}_{0.75}\text{CoO}_2$ single crystal at different temperatures. Unfortunately, this experiment suffered from the poor crystal quality. However, we observed weak incommensurate peaks at low Q , see Fig. 2, which are not of magnetic origin, as we checked by measuring the temperature dependence. The peaks are present at 10 K and remained visible at least up to 35 K. We tentatively attribute this type of Bragg reflection to a superstructure in the sodium layers.

References:

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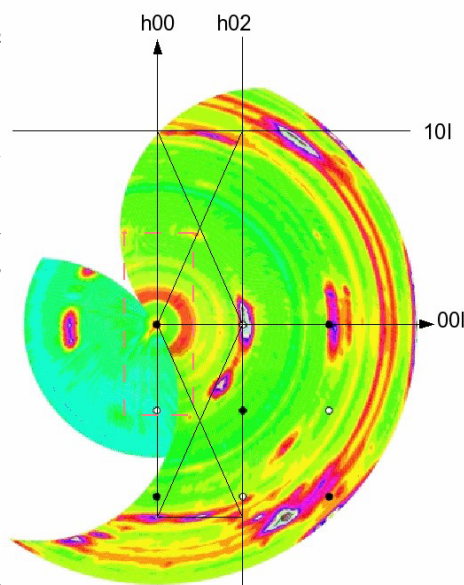


Fig. 2: Scattering by a $\text{Na}_{0.75}\text{CoO}_2$ single crystal at 10 K.



Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-04-018		
Experiment title:	A novel approach to the structure of ^1H -containing liquids: the case of liquid water		
Dates of experiment:	18-22 April, 2005	Date of report:	23 March 2006
Experimental team: Names	Addresses		
PUSZTAI, László	Research Institute for Solid State Physics and Optics, Hungarian Academy of Sciences, P.O. Box 49, Budapest, H-1525, Hungary (e-mail: lp@szfki.hu)		
SCHWEIKA, Werner			
Local Contact:	W. Schweika		

Experimental report text body

Precise knowledge of the microscopic structure of liquid water is of utmost importance for most chemists and molecular biologists - the reason, quite simply, is that life on Earth is based on water and takes place in aqueous solutions. For this reason, water has been the subject of a huge number of diffraction studies. Despite this fact, liquid water is considered to be one of the most notorious cases: for instance, there is an uncertainty of about 0.15 Å even in terms of the *position* of the first intermolecular O-H distance (which characterize hydrogen bonding - so the knowledge of this distance is crucial!) [1]. The reason for this is, to cut the long story short, *hydrogen*.

Neutron diffraction with H/D substitution seems to be the only feasible way of deriving detailed information on the microscopic structure of hydrogenous (i.e., containing ^1H) systems. However, the exceptionally high level of spin-incoherency of ^1H renders more than 95 % of the measured diffraction signal from pure H_2O nearly useless ('background') from the structural point of view (see, e.g., [2]).

Neutron diffraction with H/D isotopic substitution poses great but many times, impassable difficulties during data analyses. Strangely enough, the beneficial potential of polarised neutron scattering has not really been exploited so far in this respect (maybe because traditional areas for polarised neutrons are quite far from chemistry and biology). Among very few instruments, the DNS instrument at FZ Jülich offers the possibility polarized neutron diffraction for the wide-angle scattering of liquids.

Polarized neutron diffraction measurements have been carried out on liquid water samples containing different amounts of light and heavy water (H_2O and D_2O , respectively). For each sample, temperature scans (under atmospheric pressure) were performed, within the normal liquid range of the samples. At each temperature, both the 'spin-flip' and 'non-spin-flip' scattering were determined, so that the coherent and the

incoherent contributions to the total scattering could be separated. The coherent total structure factors were then subjected to Reverse Monte Carlo (RMC) modeling [3] studies. In the RMC calculations, thousands of flexible water molecules were applied, so that the behavior of the bulk could be mimicked.

In the Figure to the right, coherent total scattering functions are shown for 4 different H/D isotopic compositions (symbols; 'zero water' contains about 65 % of H₂O and 35 % D₂O) at room temperature. The solid curves represent the corresponding Reverse Monte Carlo structural models, which were obtained by modeling all the 4 measured functions simultaneously. As it is obvious, the models are fully consistent with results of the DNS experiments within the uncertainties of the experiments.

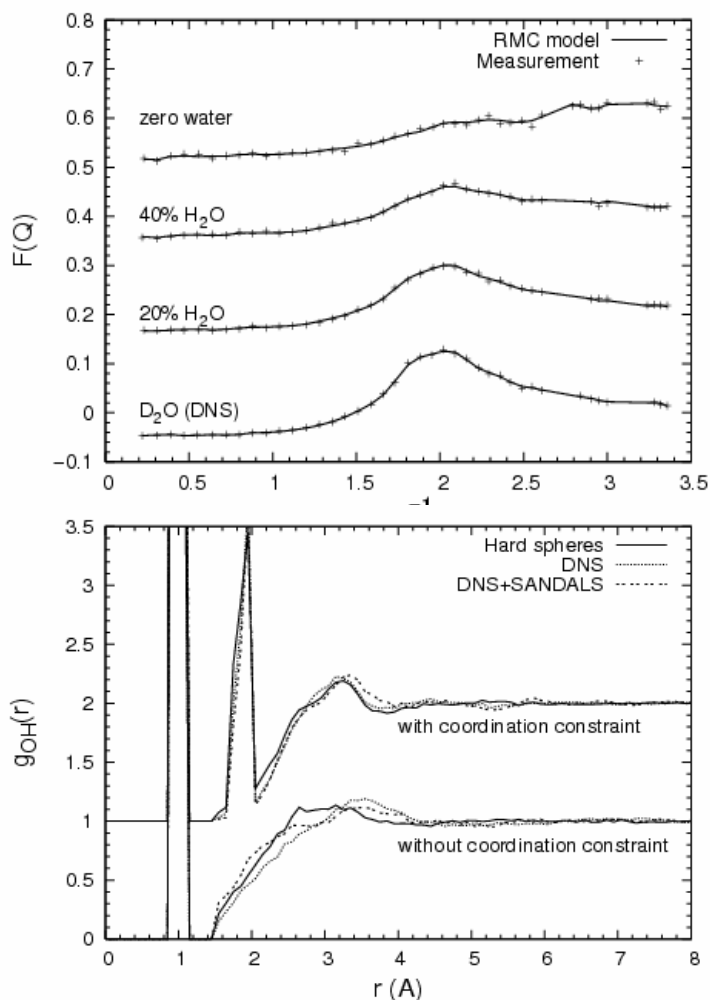
From the particle co-ordinates in the model, several characteristics of the structure could be determined. For instance, the partial radial distributions functions (prdf) can easily be calculated directly from the co-ordinates, so that the (in many cases, rather cumbersome) process of traditional separation (using inversion of the coefficient matrix) may be avoided.

In the Figure to the right, the most important of the real space partial radial distribution functions, the O-H prdf is displayed, for various models of the structure. For reference, hard sphere Monte Carlo calculations have also been carried out; in this Figure, results from these simulations are also shown. It is obvious that for a meaningful description of hydrogen bonding, co-ordination constraints (on the non-bonding O...H atom pairs) had to be implemented, in addition to the constraints already imposed by the experimental data (see upper part of the Figure). Note that all but the hard sphere O-H prdf's are consistent with DNS data, measured between 0.2 and 3.4 Å⁻¹. Addition of D₂O data measured over a much wider Q-range [4] to the set of data modeled has not changed the prdf's; contrary, the new results from DNS impose a significant further confinement of the configurational space for the modeling procedure.

In summary, it has been demonstrated that the combination of polarized neutron diffraction and RMC modeling does present a feasible way for the determination of the microscopic structure of hydrogenous materials (including our example here, water). Extending the Q-range of the measurements, however, would be rather desirable; the new DNS instrument at FRM-II (Munich) will provide us with the possibility of measuring the signal beyond 5 Å⁻¹.

References

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Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-05-004		
Experiment title:	Spin excitations in frustrated triangular antiferromagnet YMnO₃		
Dates of experiment:	29.9.05 – 7.10.05	Date of report:	December 14, 2005
Experimental team: Names	Addresses		
Tapan Chatterji W. Schweika	Institut Laue-Langevin, BP 156, 38042 Grenoble Cedex, France IFF, FZ Juelich		
Local Contact:	W. Schweika		

Experimental report text body

The hexagonal manganites RMnO₃ (R = Y, Ho, Er, Tm, Yb) are both ferroelectric and antiferromagnetic [1-6] and the corresponding order parameters couple to each other although the ferroelectric transition temperature is as high as $T_C = 900$ K whereas the Néel temperature is $T_N = 90$ K. This coupling between ferroelectric and magnetic order parameters yields the possibility of controlling electrical properties through magnetic field and vice versa and therefore hexagonal manganites may have potential applications in electronics. Due to the quasi-two dimensional triangular lattice, the magnetic system is geometrically frustrated, which is evidenced by the large ratio of Weiss-to-Néel temperature. We mapped the spin wave dispersion of YMnO₃ on the thermal triple-axis spectrometer IN3 of the Institute Laue-Langevin, Grenoble, using a multi-analyzer detector at two temperatures below and above T_N . This investigation suggested the existence of persistent spin wave above the ordering temperature. Due to the lack of beam time we confined our investigations in the (h0l) scattering plane. In the present investigation on the DNS spectrometer we wished to extend the measurements in the (hhl) plane

We have measured spin dynamics of YMnO₃ in the (hhl) scattering plane on DNS of FZ-Jülich at $T = 12, 40$ and 75 K. This scattering plane was chosen because we previously investigated the spin dynamics on the thermal triple-axis spectrometer IN3. We knew that most of the inelastic scattering appears in (h0l) plane but wished to investigate the (hhl) plane that was not investigated before. At $T = 12$ and 40 K we observed practically no magnetic signal but only scattering intensity which we attributed to the phonon scattering. At $T = 75$ K which is 5 K above T_C we however observed paramagnetic scattering along $Q = (1/2, 1/2, l)$. Fig. 1 and 2 gives the diffuse scattering at different temperatures.

References:

1. T. Katsufujii et al., Phys. Rev. B **64**, 104419 (2001).
2. Th. Jolicouer and J.C. Le Guillou, Phys. Rev. B **40**, 2727 (1989).
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6. J.M. Kosterlitz and D.J. Thouless, J. Phys. C **6**, 1181 (1973); J.M. Kosterlitz, J. Phys. C **7**, 1047 (1974)

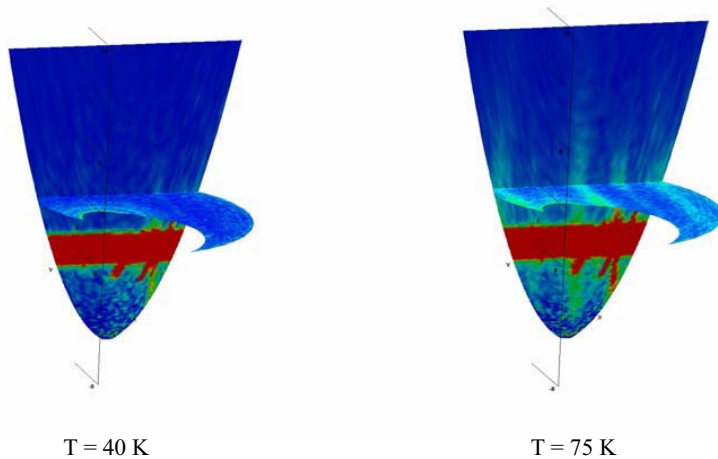


Fig. 1 – Diffuse scattering from YMnO_3 at $T = 40 \text{ K}$ and $T = 75 \text{ K}$.

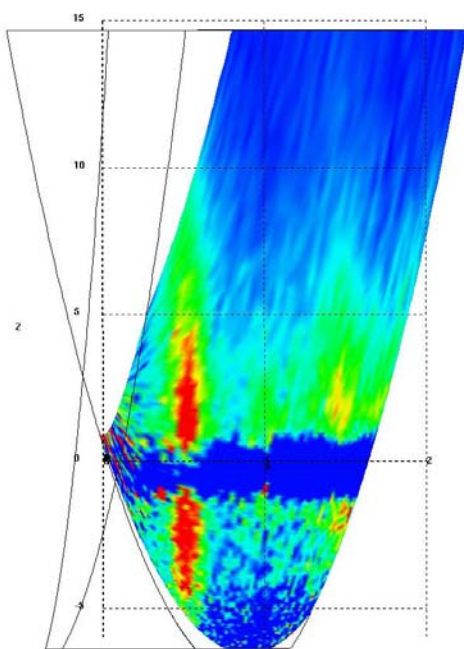


Fig. 2 – Magnetic scattering from YMnO_3 determined at $T = 75 \text{ K}$ after subtracting the background scattering measured at $T = 12 \text{ K}$.



Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-05-005		
Experiment title:	Magnetic Critical Scattering of MnO in Confined Geometry		
Dates of experiment:	10.05.05 – 21.05.05	Date of report:	31.05.06
Experimental team: Names	Addresses		
Mikhail Feygenson Dr. Werner Schweika Dr. Alexander Ioffe Prof. Thomas Brückel	Forschungszentrum Jülich GmbH Institut Für Festkörperforschung(IFF-STM) D-52425 Jülich Germany		
Local Contact:	Dr. Werner Schweika		

Experimental report text body

The aim of the experiment was to study the magnetic phase transition of MnO embedded in a porous glass. It has been claimed previously [1] that the discontinuous first order transition in the bulk becomes continuous in confined geometry. However, it is difficult to study the critical behavior because of a strong background from nuclear scattering in the glass. Therefore, we used XYZ polarization analysis at the DNS spectrometer that allow us to suppress this background by a few orders of magnitude and thus to observe a weak magnetic scattering.

The temperature dependence of the magnetic ($\frac{1}{2} \frac{1}{2} \frac{1}{2}$) Bragg peak was investigated for the wavelength of 5.24 Å within the temperature range from 2 K up to 130 K. Angular resolution was improved by a proper collimation of incident beam. As the reference, the MnO powder was investigated between 11K and 140K with the same setup. The temperature dependence of the magnetic ($\frac{1}{2} \frac{1}{2} \frac{1}{2}$) Bragg peak is shown in Fig.1. The measuring time is significantly increased comparing to the MnO powder due to the low content of MnO inside the glass. The normalized intensity for both samples obtained from the observed peaks is shown as a function of temperature in Fig.2.

It is seen that transition temperature for MnO glass is slightly increased compared to those for the MnO powder $T_N = 118$ K [2], however, the order parameter is lower for the MnO glass for most of the temperatures. The magnetic transition is second order as previously observed by Golosovsky et. al.[1]. There is a relation between pressure and ordering temperature [1,3]. According to the phase diagram given in [3] the transition temperature of MnO increases with increasing pressure. The frustration due to broken bonds at the interface can lead to the surface induced disorder as discussed in Ref.[4] and may explains why the order parameter of the MnO glass is lower compared to the bulk behavior and resembles a continuous phase transition.

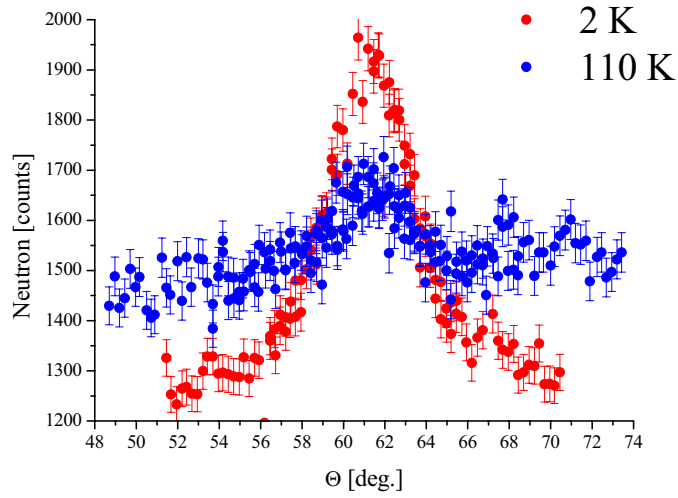


Fig.1. Magnetic ($\frac{1}{2} \frac{1}{2} \frac{1}{2}$) Bragg peak of MnO embedded in glass at 2 K and 110 K. The peak is observed in the spin flip channel and contains incoherent and magnetic scattering.

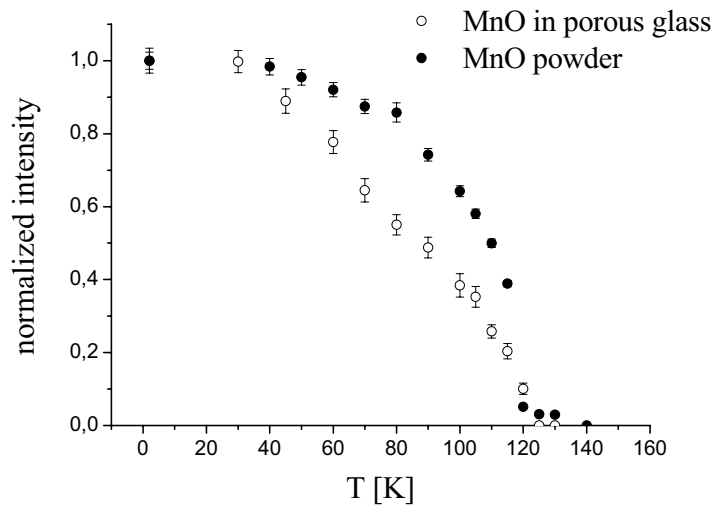


Fig.2. Normalized intensity as a function of temperature for the MnO powder embedded in a porous glass and for the MnO powder.

References:

- [1] Golosovsky et. al., PRL 86 (2001).
- [2] Lines et. al., PR. 139, 4A (1965)
- [3] Yoo et. al., PRL 94 115502 (2005)
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Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-05-006		
Experiment title:	Q-dependence of the coherent contribution to the scattering in a PEO-PMMA blend		
Dates of experiment:	27.10.2005	Date of report:	25.11.2005
Experimental team: Names	Addresses		
Katarzyna Niedzwiedz	IFF, Forschungszentrum Jülich, 52425 Jülich		
Dr. Andreas Wischniewski	IFF, Forschungszentrum Jülich, 52425 Jülich		
Dr. Michael Monkenbusch	IFF, Forschungszentrum Jülich, 52425 Jülich		
Prof. Dr. Dieter Richter	IFF, Forschungszentrum Jülich, 52425 Jülich		
Local Contact:	Dr. Werner Schweika		

Experimental report text body

The experiment was performed to determine the ratio of coherent and incoherent scattering intensities for a polymer blend with protonated PEO and deuterated PMMA.

To investigate the dynamics of the PEO chains in the blend, backscattering measurements on a mixture of protonated PEO and deuterated PMMA have been performed (proposal BSS-05-008). For a quantitative evaluation of the data one has to know how strong the incoherent (from protonated PEO) and coherent (from the deuterated PMMA and the single chain structure factor at low Q) contribution to the full scattering intensity is (see for example [1]).

For this purpose in this experiment we used the polarization analysis of the DNS spectrometer.

We have measured the ratio of coherent and incoherent scattering as a function of the momentum transfer. The wavelength of the incoming neutrons was 3.3 Å. The measurement was performed for the two different PEO / PMMA compositions as measured in the BSS-05-008 proposal: 35 % h-PEO / 65 % d-PMMA and 50 % h-PEO / 50 % d-PMMA (fig. 1).

Furthermore for the 35% h-PEO / 65% d-PMMA sample several temperatures were studied (fig. 2).

The information extracted from this experiment is an indispensable input for the data evaluation of the backscattering data.

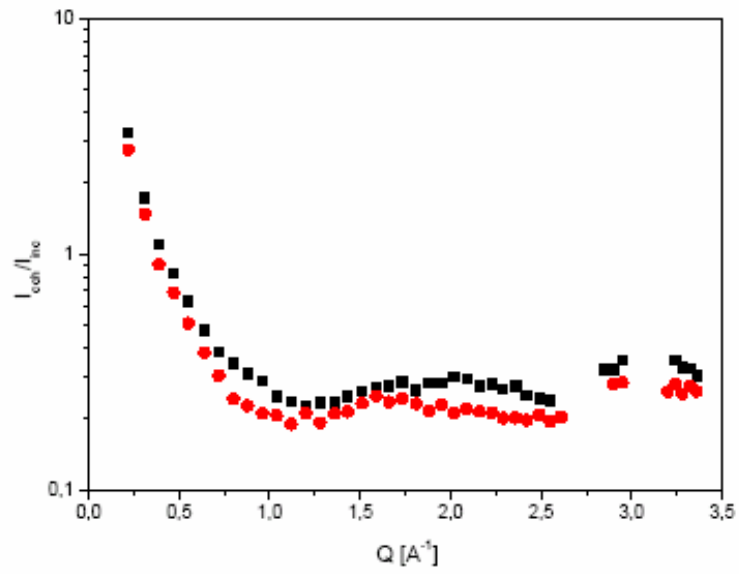


Fig. 1: Ratio between coherent and incoherent scattering intensities as a function of momentum transfer obtained for the h-PEO/d-PMMA polymer blend systems at $T = 375$ K. Black squares correspond to the 35% hPEO / 65% dPMMA sample and red circles to the 50% hPEO / 50% dPMMA sample.

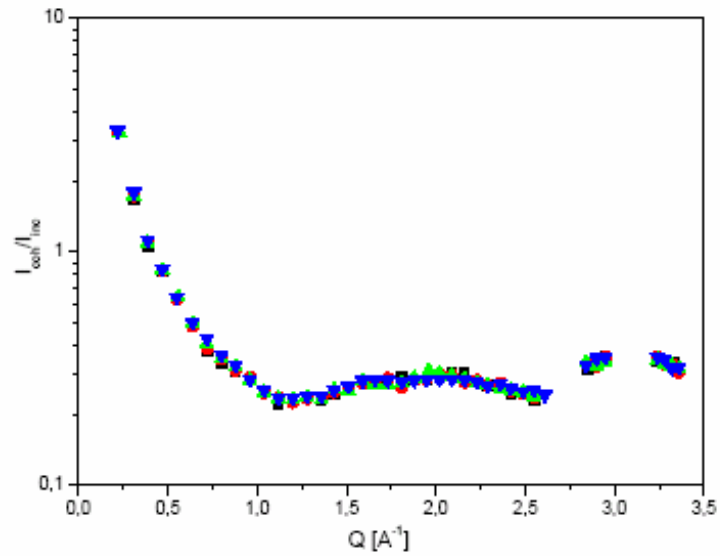


Fig. 2: Ratio between coherent and incoherent scattering intensities as a function of momentum transfer obtained for the 35% h-PEO / 65% d-PMMA polymer blend system at different temperatures: black symbols $T=350$ K, red symbols $T=375$ K, green symbols $T=400$ K and blue symbols $T=425$ K.

[1] A.-C. Genix et al., Physical Review E 72, 031808 (2005)



Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-05-010		
Experiment title:	Characterisation of a glass-forming microemulsion in the intermediate Q range		
Dates of experiment:	08.12. – 14.12.2005	Date of report:	31.05.2006
Experimental team: Names	Addresses		
M. Mayorova R. Zorn	Institut für Festkörperforschung, Forschungszentrum Jülich Institut für Festkörperforschung, Forschungszentrum Jülich		
Local Contact:	W. Schweika		

Experimental report text body

To investigate the glass transition dynamics of propylene glycol (PG) confined to reverse micelles of nanometre size three component system was chosen: propylene glycol as investigated material which was enclosed within the inversed micelles, AOT – ionic surfactant for system stabilization, decalin as oil phase ($T_g=135K$).

The composition of the microemulsion used was h-PG: 10.2% / d-AOT: 36.1% / d-decahydronaphthalene (d-decalin): 53.7% (equivalent wt% of undeuterated compounds).

An earlier SANS experiment with h-PG/h-AOT/d-decaline isotopic composition showed that no aggregation of droplets takes place. The size of the droplets could not be determined unambiguously as observed data provide information only about low q range.

To complete obtained on SANS with higher q values measurements on DNS were performed. This allows one to separate coherent and incoherent scattering so one doesn't need to make additional measurements of the ddd microemulsion for subtraction.

Table I

T, °C	Ra, E	Rhs, E	p
-24	8,621	9,875	0,22
3,6	8,610	9,220	0,1859
48,9	8,548	8,581	0,1299

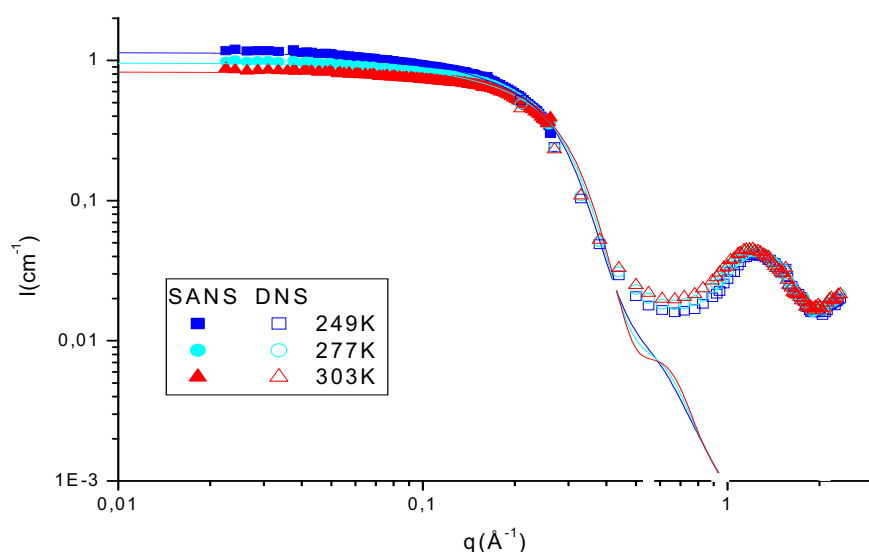


Fig. 1: SANS and DNS patterns of the h-PG/d-AOT/d-decalin microemulsion for temperatures – 24.0 ± 1 °C, $+3.6 \pm 1$ °C, and $+48.9 \pm 1$ °C. The continuous lines show a fit with the model described below.

The SANS and DNS data were combined and fitted by a model based on the following assumptions: polydispersity is small, structure function has Percus-Yevick function form (characterized by an equivalent hard-sphere radius R_{hs}) and Schultz distribution (characterized by an average radius R_a and a polydispersity p) is suitable.

As a result of this fit it was found that there is an extremely small confinement with characteristic averaged radius of PG core about 0,86 nm. It is three times lower than it was predicted from a simple geometrical assumption (the volume fractions of AOT and PG and the thickness of the AOT layer from literature [1,2]). The fit parameters shown in Table I indicate that the average radius of particles stays roughly constant while there is a slight increase in polydispersity towards low temperatures. Comparison of experimental data and fit displays that there is considerable deviation between them only at high q range where a contribution of the PG molecule structure factor, which is not included in fit, plays the main role.

Inelastic spectra for hdd at 100K and 2 K and ddd at 100K samples were also obtained on DNS. They will be combined with data from Focus spectrometer to get the vibration density of states function which contains information about Boson peak, a deviation from linear behavior of density of states function due to an excess of states connected to the existence of localized soft modes.

- [1] Lin-Min Wang, Fang He, R. Richert, *Phys. Rev. Lett.* **92**, 095701 (2004)
- [2] M. Kotlarchyk, S.-H. Chen, J. S. Huang, M. W. Kim, *Phys. Rev. Lett.* **53**, 941 (1984)



Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-05-012		
Experiment title:	Magnetic disorder and transitions in the chemical system (Y,Ca)BaCo ₄ O ₇		
Dates of experiment:	7-13.11.2005	Date of report:	24.3.2006
Experimental team: Names	Addresses		
Martin Valldor	Institut für Physik der Kondensierten Materie, TU-Braunschweig, Mendelssohnstrasse 3, D-38106 Braunschweig, Germany Institut für Physik der Kondensierten Materie, TU-Braunschweig, Mendelssohnstrasse 3, D-38106 Braunschweig, Germany Institut für Streumethoden, IFF, Forschungszentrum Jülich, D-52425 Jülich, Germany		
Peter Lemmens			
Werner Schweika			
Local Contact:	Werner Schweika		

Experimental report text body

The two samples YBaCo₄O₇ and Y_{0.5}Ca_{0.5}BaCo₄O₇ were investigated using polarised neutrons to separate magnetic from nuclear scattering and data was collected at several different temperatures between 1.2 and 440 K. For YBaCo₄O₇, a magnetic order is seen below the postulated transition temperature (65 K) [1] as a sharp magnetic intensity (at 30 K) close to one of the nuclear Bragg-planes (101 ~ 1.3 Å⁻¹) (Fig.1). The sharpness of the magnetic peak indicates long-ranged order, but there is also a broad magnetic intensity with its maximum about 1.6 Å⁻¹, suggesting that parts of the spins still are disordered or fluctuating. At higher temperatures (100 K), the magnetic scattering is smeared out, but is not completely gone, indicating fluctuating short-range magnetic order. This is a further proof of extremely strong magnetic frustration, which was already seen in the magnetic susceptibility as a Curie-Weiss constant of about -1000 K [1]. At 300 K, The spins are fully decoupled and little structure is seen in the magnetic scattering apart from the average scattering factor of Co.

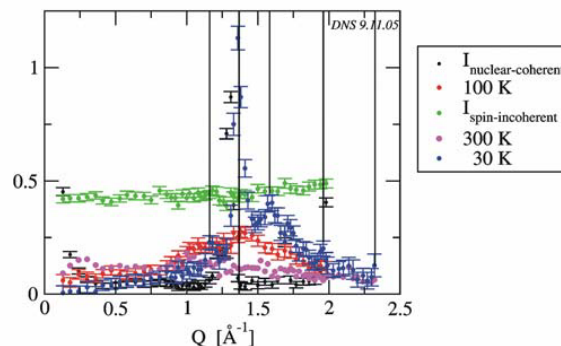


Fig.1 Neutron nuclear and magnetic scattering on YBaCo₄O₇ at three different temperatures.

$Y_{0.5}Ca_{0.5}BaCo_4O_7$ exhibits a totally different scattering behaviour although the only difference is that half of the Y has been substituted for Ca. This Ca substitution results in an increase in Co valence with drastic consequences for the magnetic coupling between the Co ions: no long-ranged order can be detected down to 1.2 K, but a broad intensity is observed with its maximum close to $1.33 \text{ \AA}^{-1}$ (Fig.2). The lack of sharp magnetic peaks can only be explained by the fact that the chemical doping by Ca has cancelled the magnetic order, leaving a strongly frustrated magnetic lattice.

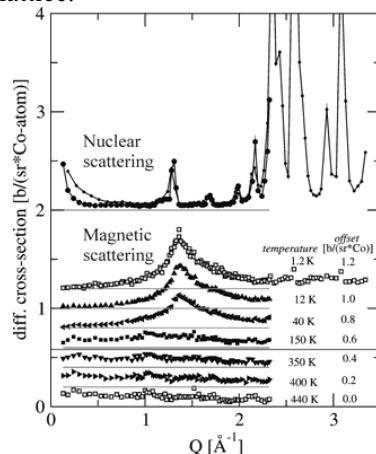


Fig.2 Neutron nuclear and magnetic scattering on $Y_{0.5}Ca_{0.5}BaCo_4O_7$ at several different temperatures; the magnetic scattering is drawn versus Q below the nuclear scattering; temperatures are indicated to the right of the data. The data have been shifted with an offset for comparisons.

The asymmetric peak shape indicates two-dimensional (2D) antiferromagnetic (AF) spin correlations, which are well described by theoretical predictions of Reimers and Berlinsky[2] for the 2D kagome AF with only nearest neighbor interactions. The spin correlations resemble the so-called $\sqrt{3} \times \sqrt{3}$ spin structure (Fig.3) for the Co atoms on the kagome layers in $Y_{0.5}Ca_{0.5}BaCo_4O_7$. The absence of an AF long range ordered state, in accordance with the Mermin-Wagner theorem, confirms the lack of any noticeable interlayer coupling as found in the un-doped material. When lowering the temperature, a peak of the magnetic diffuse intensity evolves at $1.33 \text{ \AA}^{-1}$, which perfectly agrees with the ordering wave vector of the $\sqrt{3} \times \sqrt{3}$ spin structure for the cell parameter of $a=6.3 \text{ \AA}$ of $Y_{0.5}Ca_{0.5}BaCo_4O_7$ (Fig.2). Further studies on similar systems, also with polarised neutrons, are on the way.

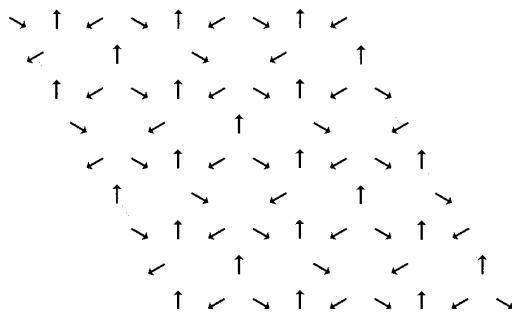


Fig.3 The in-plane spin motif on the kagome lattice of Co_2 , presented as lines where the crossing points are the sites for Co_2 . This spin structure is called $\sqrt{3} \times \sqrt{3}$ [2].

[1] Valldor, M. & Andersson, M. The structure of the new compound $YBaCo_4O_7$ with a magnetic feature. *Solid State Sci.* **4**, 923-931 (2002)

[2] Reimers, J.N. & Berlinsky, A.J. Order by disorder in the classical Heisenberg kagomé antiferromagnet. *Phys. Rev. B* **48**, 9539-9554 (1993).



Experimental Report of Neutron Scattering Experiments at the FRJ-2 Reactor

Proposal number:	DNS-05-013		
Experiment title:	High Spin – Low Spin transition in per deuterated $\text{Fe}(\text{NCS})_2(\text{brt})_2\cdot\text{D}_2\text{O}$. The magnetic inelastic response as determined by xyz-PNA.		
Dates of experiment:	17.02. – 22. 02.2006	Date of report:	29.06.2006
Experimental team: Names	Addresses		
D.Visser W.Schweika	NWO-Physics, CCLRC ISIS Facility, Chilton Didcot, OX11 0QX, UK Institut für Festkörperforschung, Forschungszentrum Jülich GmbH D-52425 Jülich, Germany		
Local Contact:	Dr Werner Schweika		

Experimental report text body

Spin-crossover solids are materials which are studied for future applications of optical memories or numerical displays. These materials are also good examples of photo excitable materials. Optical switching at low-temperatures, below the high spin - low spin transition ($T(\text{hs-ls})$) is achieved by the so-called direct or inverse LIESTT effect (Light Induced Excited spin State Trapping) [1, 2].

The structurally quasi two-dimensional molecular magnet $\{\text{Fe}(\text{NCS})_2(\text{brt})_2\}\cdot\text{H}_2\text{O}(\text{D}_2\text{O})$ was the first material reported to exhibit a high spin (HS) to low spin transition (LS) around the $T_{\downarrow}(\text{hs-ls}) = 124\text{K}$ with a hysteresis effect as wide as 20K on increasing the temperature: $T_{\uparrow}(\text{hs-ls}) = 144\text{K}$. Since the Fe ions are co-ordinated via bi-triazole bridges, the effect of HS - LS transition is dramatic. The high temperature paramagnetic behaviour changes abruptly to a low temperature diamagnetic one at $T(\text{HS} - \text{LS})$ via a first-order structural phase transition. This transition is also accompanied with a colour change from transparent to purple. This transition has been observed by magnetisation, specific heat, Mössbauer and optical spectroscopy [3]. Recently we studied this transition with muon spin resonance [4]. In the diamagnetic phase one would have expected a silence in the muon relaxation behaviour. However, a complex relaxation behaviour is observed which was tentatively attributed to muonium formation but no hard evidence was obtained for a non-magnetic ground state in the low-temperature phase.

To obtain further information about the magnetic character of the $\{\text{Fe}(\text{NCS})_2(\text{brt})_2\}(\text{D}_2\text{O})$, we determined the inelastic response using xyz-polarisation analysis at $T = 160\text{ K}$ (high spin state) and $T = 190\text{ K}$ (low spin state) using the high resolution set up on the DNS polarised neutron diffractometer. We performed TOF measurements on a powder sample of fresh fully deuterated $\{\text{Fe}(\text{NCS})_2(\text{brt})_2\}\cdot(\text{D}_2\text{O})$ sample. At $T = 190\text{ K}$, we observe two excitation levels in the energy loss side of the inelastic neutron response: 0.8 meV (as a shoulder near the elastic line) and 1.6 meV, see fig.1. These excitations are absent in the 90 K data, see fig.2. The difference spectrum of both data sets show both excitations clearly. Independently the magnetic signal can be observed in

the difference spectrum from the $I_{y,sf} - I_{y,nsf}$ signal., see fig 3.

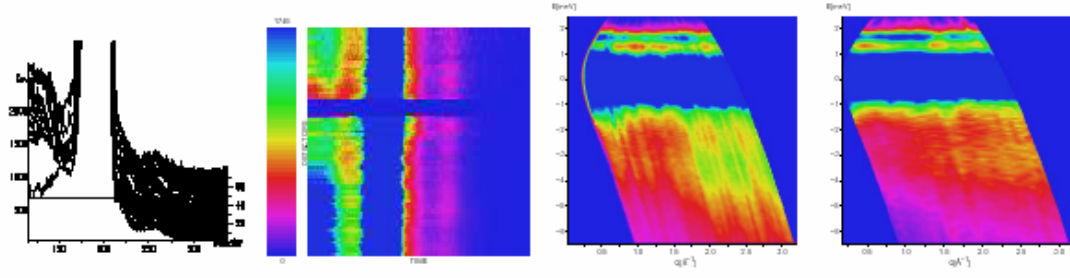


FIG. 1: T=160K, sum of all data, background corrected

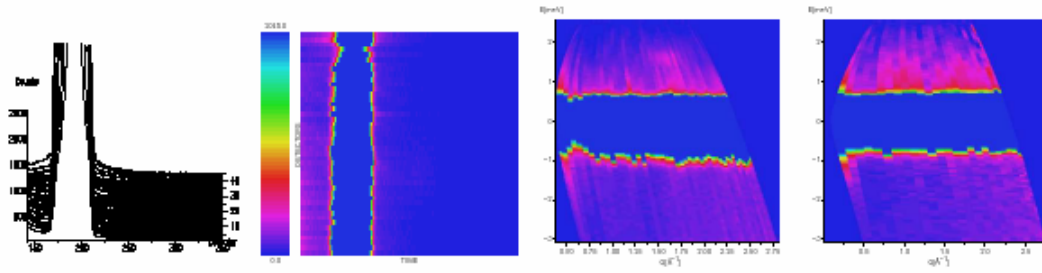


FIG. 2: T=90K, sum of all data, background corrected

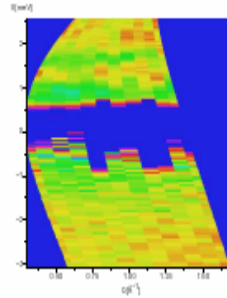


FIG. 4: T=160K. $I_{y,sf} - I_{y,nsf}$. A proof of the magnetic nature of these excitations is in principle possible using the xyz-method. However, the statistical quality is poor for such differences. An indication for the magnetic nature is found by taking differences [$x_{sf} - x_{nsf}$ perp Q almost no signal and $z_{sf} - z_{nsf}$ $\perp$ Q almost no signal] but for $y_{sf} - y_{nsf}$ $\parallel$ Q, there is a signal, see figure above.

The magnetic excitations are dispersionless. This indicates that we are looking at crystal field levels instead of spinwave like excitations. Further analysis has to show if the intensity of the excitations are Q-dependent, which would mean that they would not be a single spin excitations. we have shown that the ground state of $\{\text{Fe}(\text{NCS})_2(\text{brt})_2\}(\text{D}_2\text{O})$ is non-magnetic.

References:

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- [2] O.Kahn et al. Science 279 (1998) 44.
- [3] W. Vreugdenhil et al. Polyhedron 9 (1990) 2971.
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Experimental Report
of Neutron Scattering Experiments
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Proposal number:	DNS-05-014		
Experiment title:	The correlations between the ferroelectric and magnetic transitions in REMn_2O_5		
Dates of experiment:	01.12.2005	Date of report:	01.06.2006
Experimental team: Names	Addresses		
Lieh-Jeng Chang	Nuclear Science and Technology Development Center National Tsing Hwa University 101, Sec. 2, Kuang-Fu Road HsinChu, 30013, Taiwan		
Local Contact:	Dr. Yixi Su		

Experimental report text body

Introduction

There is currently a great interest in the materials that exhibit interplay between lattice distortions and electrical and magnetic ordering. Among these materials, magnetoelectrics, in which magnetism and ferroelectricity coexist and are mutually coupled, are being investigated extensively. These materials display phenomena such as the control of electrical polarization by the application of an external magnetic field, providing an additional degree of freedom for the design of new devices.¹ However, compounds showing large magnetoelectric effects are extremely rare. The magnetoelectric TbMn_2O_5 exhibits a profound interplay between electrical polarization and the applied magnetic field. This is demonstrated by two discoveries: the highly reversible switching of electrical polarization using relatively low magnetic fields of 0-2T; and the combined application of electric and magnetic field, which leaves a permanent imprint in the polarization.²

TbMn_2O_5 is an insulator and consist of linked Mn^{4+}O_6 octahedra and Mn^{3+}O_5 pyramids adopting space group $Pbam$. The octahedral share edges to form ribbons parallel to the c axis, adjacent ribbons being linked by pairs of corner-sharing pyramids.¹ The structure at room temperature is orthorhombic³. The reported neutron diffraction measurements on TbMn_2O_5 as a function of temperature and applied field which show unambiguous correlations between dielectric anomalies and changes in the periodicity of the spin structure. First, the onset of ferroelectricity at 38 K is associated with the appearance of incommensurate antiferromagnetic ordering $T_1 = 43$ K. At $T_2 = 33$ K, just above the maximum in spontaneous polarization $\mathbf{P}$, propagation vector $\mathbf{k}$ locks into a commensurate value $(1/2, 0, 1/4)$. Remarkably, at $T_3 = 24$ K, where the dielectric constant ϵ has an upward “jump”, $\mathbf{k}$ becomes suddenly incommensurate again, a rare example of such a transition on cooling. The low temperature transition ($T_4 = 10$ K) coincides with a major increase of the Tb ordered moment. The 43 and 24 K transitions are accompanied by clear anomalies in the lattice parameters, particularly along the b “polar” axis, and in the atomic displacement parameters.⁴

Experiments

8 grams of TbMn_2O_5 polycrystalline sample have been prepared through conventional solid-state reaction method and characterized by powder x-ray diffraction and magnetization. All the observable peaks on the powder x-ray pattern can be well indexed for showing the sample to be single phase. A broad peak has been observed on the magnetic susceptibility from 50 to 20 K corresponding to the series of transitions mentioned above. Experiments were performed at DNS in an orange cryostat for temperatures between 1.2 K and 250 K. Neutron wavelength 4.8 Å was selected for the elastic scattering experiments. Neutron polarization analysis was also performed to separate the nuclear and magnetic scatterings.

Results

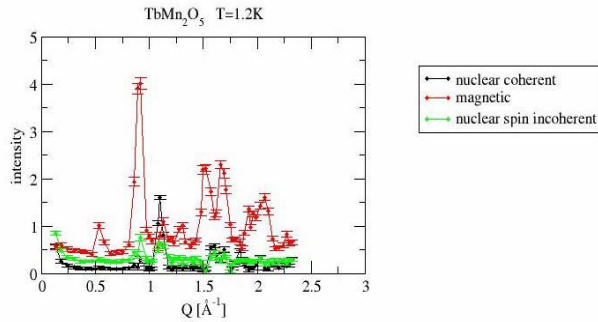


Fig. 1 Elastic scattering from DNS using neutron wavelength 4.8 Å at temperature 1.2 K with neutron polarization analysis. The nuclear spin incoherent peak at $Q \sim 0.8\text{ \AA}^{-1}$ is thought not from nuclear but of magnetic origin due to insufficient separation. This may be due to either not perfect corrections for flipping ratio background and multiple scattering and / or it may have a cause also in the complex non-collinear structure.

Fig. 1 shows the elastic spectra using the neutron wavelength 4.8 Å taken at 1.2 K with neutron polarization analysis. The nuclear spin incoherent peak at $Q \sim 0.8\text{ \AA}^{-1}$ is thought not from nuclear but of magnetic origin due to insufficient separation. This may be due to either not perfect corrections for flipping ratio background and multiple scattering and / or it may have a cause also in the complex non-collinear structure. The high magnetic background reveals that the magnetic moments have not ordered completely even down to 1.2 K.

Fig. 2 shows Magnetic peaks observed at different temperatures. Long-range ordered magnetic peaks are observed from $\sim 39\text{ K}$. This is consistent to the published results that the antiferromagnetic order occurs at 43 K.⁴ The background at 250 K is lower than that of 48 K and 60 K is that at high T the width of the paramagnetic scattering will be about 250 K $\sim 21.5\text{ meV}$. This means that the transmission of the polarisation analysers, which are less efficient for faster neutrons, have to take into account This also means that only 60 to 50 % of the total paramagnetic scattering at high T have been observed. To clarify it, time-of-flight measurements and a more detailed analysis will be useful to quantify the whole contribution.

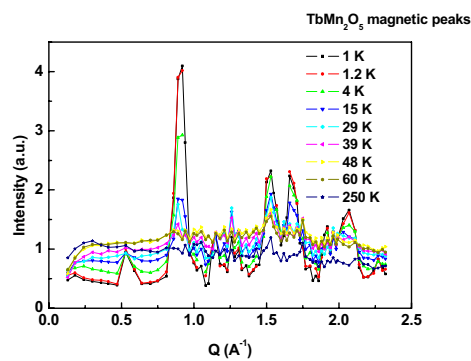


Fig. Magnetic peaks observed at different temperatures. Long-range ordered magnetic peaks are observed from $\sim 39\text{ K}$. This is consistent to the published results that the antiferromagnetic order occurs at 43 K.⁴

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1. G.R. Blake, et al., Phys. Rev. B **71**, 214402 (1995).
2. N. Hur, et al., Nature (London) **429**, 392 (2004).
3. D. Higashiyama and S. Miyasaka, Phys. Rev. B **72**, 064421 (2005).
4. L.C. Chapon et al., Phys. Rev. Lett **93**, 177402 (2004).



Experimental Report
of Neutron Scattering Experiments
at the FRJ-2 Reactor

Proposal number:	DNS-05-015		
Experiment title:	Q - dependence of the coherent / incoherent contribution to the scattering of 20 % PEO- 80% Montmorillonite		
Dates of experiment:	29.11.2005	Date of report:	09.03.2006
Experimental team: Names	Addresses		
Dr. Andreas Wischnewski	IFF, Forschungszentrum Jülich, 52425 Jülich		
Katarzyna Niedzwiedz	IFF, Forschungszentrum Jülich, 52425 Jülich		
Dr. Helen Hermes	IFF, Forschungszentrum Jülich, 52425 Jülich		
Dr. Michaela Zamponi	IFF, Forschungszentrum Jülich, 52425 Jülich		
Dr. Michael Monkenbusch	IFF, Forschungszentrum Jülich, 52425 Jülich		
Prof. Dr. Dieter Richter	IFF, Forschungszentrum Jülich, 52425 Jülich		
Local Contact:	Dr. Werner Schweika		

Experimental report text body

The experiment was performed to determine the ratio of coherent and incoherent scattering intensities for a mixture of 20% protonated PEO in 80% montmorillonite.

We have recently used solution intercalation to prepare composites of poly(ethylene oxide) (PEO) and sodium montmorillonite (MMT). Initial small angle neutron scattering experiments have been performed and the data described successfully¹. X-ray scattering and differential scanning calorimetry measurements have confirmed that using this sample preparation method and a suitable composition, it is possible for all the PEO to be intercalated between the silicate layers. The thickness of the intercalated PEO layers is 0.815 nm and extremely constant. Thus, this system is ideal for investigating the effect of extreme confinement on the dynamics and conformation of PEO polymer chains.

In a BSS experiment we have examined the same sample with 80% PEO content at temperatures between 375 and 425 K (proposal BSS-05-018). We found that the PEO – dynamics is in the BSS energy window and first indications of the effect of confinement in montmorillonite on the PEO dynamics have been seen. These results are promising, however, for a proper correction of the backscattering data of this system the information about the Q-dependence of the coherent / incoherent contribution to the structure factor is of utmost importance.

For this purpose the polarization analysis provided by the DNS spectrometer has been used. We have measured the ratio of coherent and incoherent scattering as a function of the momentum transfer. The wavelength of the incoming neutrons was 3.3 Å.

The information extracted from this experiment is an indispensable input for the data evaluation of the above mentioned backscattering experiment.

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Experimental Report of Neutron Scattering Experiments at the FRJ-2 Reactor

Proposal number:	DNS-05-016		
Experiment title:	Magnetic correlations in the Kagomé system (Y,Ca)BaCo ₃ (Al,Zn)O ₇		
Dates of experiment:	12.01. – 22.01.2006	Date of report:	06.06.2006
Experimental team:			
Names	Addresses		
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Experimental report text body

We studied the diffuse magnetic scattering from CaBaCo₃AlO₇, YBaCo₃ZnO₇, and CaBaCo₃ZnO₇ at several temperatures between 1.2 K and 300 K using the DNS instrument with polarised neutrons and xyz-polarisation analysis to separate magnetic from nuclear scattering.

A first results is that CaBaCo₃AlO₇ is not isostructural with the other compounds as previously reported [1], although the earlier data were based on single crystal X-ray diffractometry. Instead of the hexagonal unit cell, an orthorhombic has to be used to explain the nuclear scattering pattern from CaBaCo₃AlO₇, as can be seen in Fig.1. The orthorhombic cell is realised as the C-centring is broken and, since this is only obvious in neutron scattering data, the symmetry breaking is caused by changes in the oxygen substructure. However, due to low resolution in the obtained data, refinement of the more correct atomic structure of CaBaCo₃AlO₇ was not tried, but is planned for future investigations.

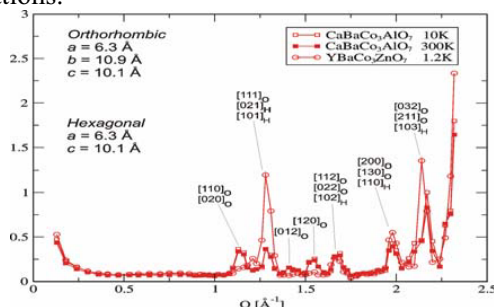


Fig.1 Comparison between nuclear scattering from the orthorhombic CaBaCo₃AlO₇ and the hexagonal YBaCo₃ZnO₇ as function of Q . Approximate unit cell parameters and indexed peaks are presented in the graph. The measuring temperatures are samples are indicated with different markers.

We do not observe any magnetic ordering transition for $\text{CaBaCo}_3\text{AlO}_7$ and $\text{CaBaCo}_3\text{ZnO}_7$ down to a temperature of 1.2 K. This is surprising in view of the strong AF-coupling expected from the high negative Curie-Weiss constants (in between -500 and -1000 K)[2], and can be taken as a sign of highly frustrated spins and/or low dimensional spin systems. Indeed, the shape of the diffuse scattering is dominated by antiferro spin correlations. A broad diffuse peak evolves in the magnetic scattering of $\text{CaBaCo}_3\text{AlO}_7$ near $1.3 \text{ \AA}^{-1}$ as shown in Fig.2a, while a more pronounced diffuse peak is found for $\text{CaBaCo}_3\text{ZnO}_7$, see Fig.2b.

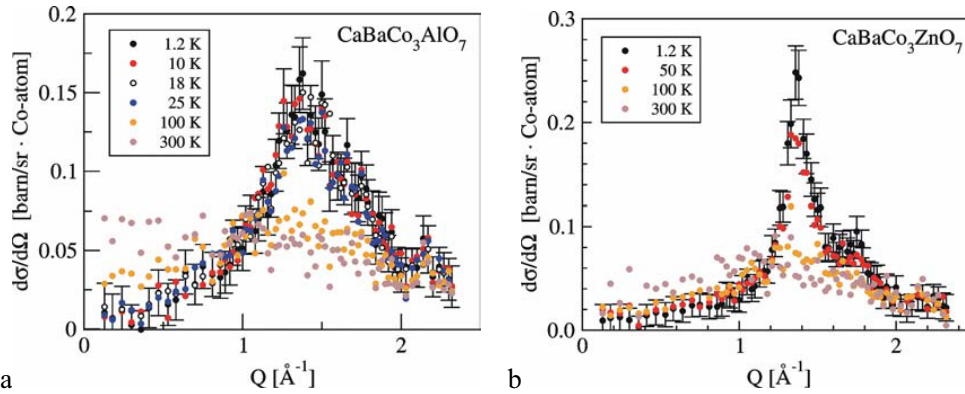


Fig.2 Magnetic scattering on $\text{CaBaCo}_3\text{AlO}_7$ (left) and $\text{CaBaCo}_3\text{ZnO}_7$ (right) at different temperatures as function of Q .

With respect to the atomic structure, the Co-sublattice can be viewed as kagomé layers which are weakly linked via another (trigonal) Co-layer. Therefore, one may expect in both compounds 2D AF spin correlations of Heisenberg spins, which would also explain the lack of Néel order down to 1.2 K. For the 2D kagomé AF with only nearest neighbor interactions, Reimers and Berlinsky[3] predicted an ordered ground state with staggered chirality, the so called $\sqrt{3} \times \sqrt{3}$ structure (Fig.3). The simulated short-range spin correlations at low but finite temperatures resemble our scattering data, with a diffuse peak close to $1.35 \text{ \AA}^{-1}$. In $\text{CaBaCo}_3\text{AlO}_7$, it is easy to explain the lack of inter-planar coupling: Al sits between the kagomé layers and hinders any magnetic coupling. The Zn substituted compound has randomly distributed Zn ions in the kagomé layers and the magnetic diffraction contains more structure: a shoulder is visible on the high Q side of the maximum diffuse scattering. The spin dilution in the kagomé layers due to Zn and possible coupling between the kagomé layers leads to a more complex situation, which has to be analysed in more detail in future.

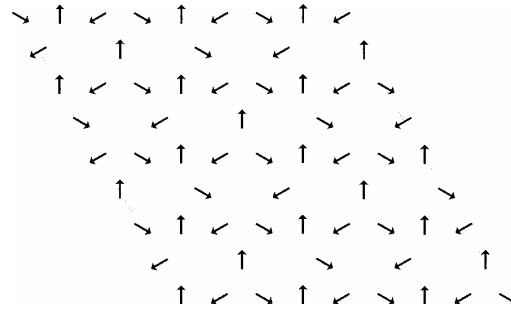


Fig.3 The in-plane spin motif on the kagome lattice of Co_2 , presented as lines where the crossing points are the sites for Co_2 . This spin structure is called the $\sqrt{3} \times \sqrt{3}$ structure[3].

- [1] Valldor M., *Solid State Sciences* 6 (2004) 251-266
- [2] Valldor M., *J. Phys. Chem. Solids* 66 (2005) 1025-1033
- [3] Reimers, J.N. & Berlinsky, A.J. *Phys. Rev. B* **48**, 9539-9554 (1993).

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Experimental Report of Neutron Scattering Experiments at the FRJ-2 Reactor

Proposal number:	DNS-05-017		
Experiment title:	Polarised neutron diffraction from multiferroic TbMnO ₃		
Dates of experiment:	9.1.06-12.1.06	Date of report:	23.5.06
Experimental team:			
Names:	Addresses:		
J. Voigt	Forschungszentrum Jülich		
Local contact:	W. Schweika		

We have studied the powder diffraction from TbMnO₃ with XYZ polarization analysis. The compound has attracted considerable interest because it exhibits multiferroic behaviour. Antiferromagnetic order and the ferroelectric polarization are intimately connected as evidenced by the polarization flop from the c -direction to the b -direction in an applied magnetic field of several Tesla [1]. The formation of the ferroelectric polarization at $T_c = 28$ K seems to be coupled to a magnetic reorientation transition from a collinear incommensurate (IC) magnetic structure to a non-collinear. This transitions involves also a long range ordering of the Tb³⁺ ions according to a recent neutron diffraction study [2]. The transition temperature is significantly higher than temperature $T_{N,Tb} = 8$ K, at which the Tb moments order with a different propagation vector as compared to the ordering of the Mn subsystem.

To resolve the magnetic structure, polarised neutron scattering give the full information about the vectors of the magnetic moments. A recent study of TbMn₂O₅ has raised the question, whether it is possible to distinguish collinear from non collinear magnetic structures. Supposedely the separation of nuclear, magnetic and spin-incoherent scattering breaks down, if the magnetic structure becomes non-collinear.

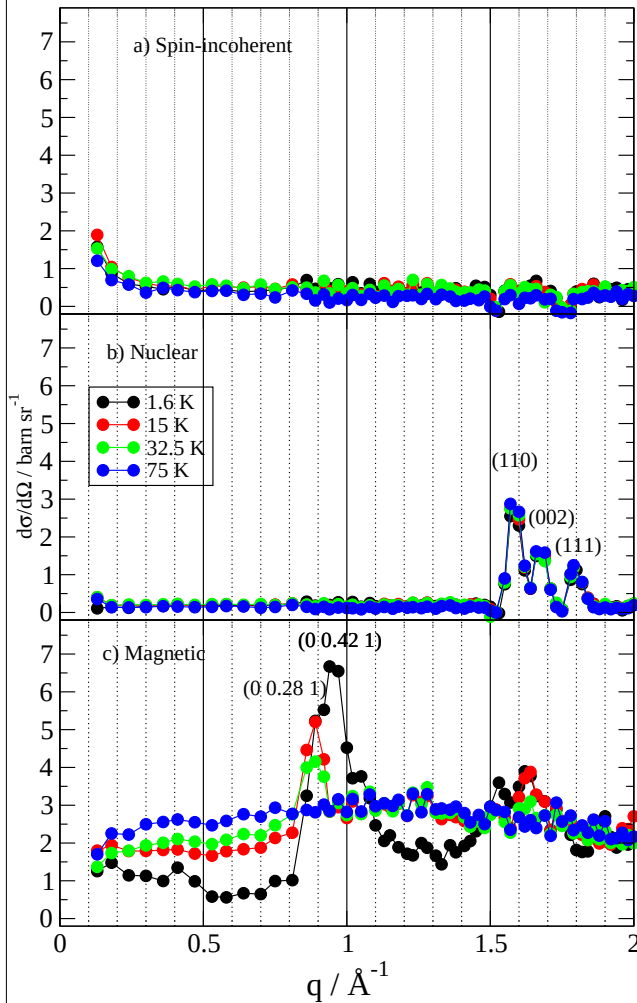
The 3.6 g of TbMnO₃ powder were mounted on DNS in a LH cryostat. Using the polarization setup, the scattering was recorded at 4 temperatures (1.6 K, 15 K, 35 K, 75 K) in the different phases identified in earlier studies, e.g [1]. For each temperature the spin-flip and non-spin-flip was collected, preparing the spin X, Y and Z direction respectively. The wavelength during the entire experiment was 4.75 Å. The different contributions shown in Fig. 1 were separated using the formulas given in [3] and calibrated by scattering from Vanadium.

Except for the small angle region and close to the structural Bragg reflections between 1.5 and 2 $\text{\AA}^{-1}$, the separation works well at all temperatures. Close to the Bragg reflection and in forward direction the separation is not perfect. The forward direction can be explained by the fact, that the neutron beam is larger than the sample.

The different contributions of the Tb IC structure ($\vec{Q}=(0\ 0.42\ 1)$) and the Mn IC ($\vec{Q}=(0\ 0.28\ 1)$) structure overlap at $T = 1.6\text{ K}$. They agree with the results from [2]. The paramagnetic scattering is also significantly weaker at this temperature.

With increasing temperature, the IC Tb structure vanishes. The transition into the paraelectric state does not lead to a significant change in any of the different contributions. The paramagnetic scattering at small momentum transfer is increased, while the magnetic peak intensity decreases. In the paramagnetic state finally the magnetic intensity is fairly broad.

Even at $T=1.6\text{K}$ there is still a significant degree of magnetic disorder. The slight increase towards small q indicates short-range ferro-type correlations within the AF-IC structure. There are no indications of long-range ferromagnetic correlations neither from magnetic scattering at a nuclear Bragg peaks nor from any loss in polarisation of the scattered beam.



References

- [1] T. Kimura *et al.*, PHYSICAL REVIEW B **71**, 224425 (2005).
- [2] M. Kenzelmann *et al.*, PHYSICAL REVIEW LETTERS **95**, 087206 (2005).
- [3] W. Schweika, in *Polarized Neutron scattering*, edited by Th. Brückel and W. Schweika (Forschungszentrum Jülich, Jülich, 2002), Chap. 1, pp. 5–29.

Fig 1: Spin-incoherent, nuclear and magnetic scattering separated by XYZ polarization analysis at different temperatures.