ILL RESEARCH PROPOSAL

Printed : 29/09/2006

Experiment Title : **Change of magnetic correlations acr oss charge ordering transtions in m anganites from polarized neutron po wder diffraction**	Proposal Number **5-53-130**

Proposer (to whom correspondence will be addressed)

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Local contact(s) : **DEEN PASCALE**

Suggested keyword number **5-53**

This proposal is :

[**x**] A new proposal.
[ ] A continuation proposal.
[ ] A resubmission.

The main research area of your proposal is

[ ] Biology [ ] Chemistry [**x**] Physics [ ] Materials [ ] Methods and instrumentation
[ ] Engineering [ ] Soft condensed matter [ ] Other :

Industry : **NOT Related to industrial applications**

Instrument required	Days
D7	**7**

Requested starting time :

1. Jan/Feb 2. Mar/Apr 3. May/Jun
4. Jul/Aug 5. Sep/Oct 6. Nov/Dec

Unacceptable Dates :

Sample availability **November 2006**

Sample description

Substance/Formula : **YBaMn2O6**
Y1/2Ca1/2MnO3

Mass (in mg) : **7-14g**

Size (in mm3) :

State : **powder**

Surface area :

Space group : **Pbnm**

Unit cell dimension : a = **5.4** b = **5.8** c = **7.6**

T (k) = **RT** α = **90** β = **90** γ = **90**

Sample container :

Safety aspects

The sample is : **TOXIC**

Is there any danger associated with the proposed sample or its preparation at ILL ?

[ ] Yes [ ] Uncertain [**X**] No If Yes or Uncertain, please give details of the risks asociated :

Is the sample a transuranium sample [ ] Yes [**X**] No

Experimental details

Energy / wavelength range : **4.8A**

Resolution in energy or wavelength :

Range of momentum transfer :

Resolution in momentum transfer :

Sample environment equipement (supplied by ILL)

Environment equipment : **furnaces 200 - 1100 C** **furnaces T>1600 C**

Temperature range (stability) : **300K<T<1000K** Pressure range :

Magnetic-field strength (stability)

Is there any danger associated with ancillary equipement ? [ ] Yes [ ] Uncertain [**X**] No
If Yes or Uncertain, please give details of the risks associated :

To be filled in by ILL

Sample environment code	Comments by Health Physics Officer and Safety Engineer
F1 F3	

Abstract

Half-doped R1/2Ca1/2MnO3 manganites (R:Y, Pr, Nd…) represent the prototype example of copmpound presenting ionic Mn3+/Mn4+ charge ordering, which seems to hold also for novel A-site ordered RBaMn2O6 materials. However, novel pictures for their ground state are emerging. In particular, Zener Polaron ordering, suggests the localization of the charge inside ferromagnetic polarons of different size in the two materials. The change of magnetic correlations across the charge ordering transition is expected to be distinguishable for the two models of charge localization, as well as it should be different for the two compounds. We intend to evidence the differences, and characterize such correlations by extracting the pure magnetic scattering cross sections by means of polarized neutron powder diffraction on the D7 spectrometer.

Scientific background

Charge ordering in the manganites is ubiquitous and influences the properties of most of them. Even colossal magnetoresistance is currently understood as electronic phase separation involving charge ordered nano-domains. But opinion on the exact nature of the ordering is sharply divided.

Long range charge ordering occurs both in prototype $R_{½}Ca_{½}MnO_3$ manganites (R:Y, Pr, Nd, La…) and less conventional A-site ordered $RBaMn_2O_6$ (R: Tb, Y) compounds. The original views about the nature of the ground state of $R_{½}Ca_{½}MnO_3$ were essentially based on a super exchange picture [1], which assumed a celebrated model of ionic charge and orbital ordering of Mn^{3+}/Mn^{4+} ions to explain their complex *collinear* magnetic structure. They have been adapted to the different ground state of $RBaMn_2O_6$ compounds [2], which is described in terms of variants of the charge/orbital orderings associated again to a *collinear* spin order, as shown in Fig.1a.

The ionic model has been addressed for years in $R_{½}Ca_{½}MnO_3$ by crystallographic and spectroscopic techniques, which first have corroborated [2], but then discarded [3,4] it. An emerging alternative would be the ordering of two manganese ions into a ferromagnetic dimer state known as a Zener polaron [3]. The proposal was deduced from structural refinements of single crystal neutron diffraction data on $Pr_{0.6}Ca_{0.4}MnO_3$ [3], but doubts on the validity of the structure supporting this interpretation were recently raised by resonant X-ray scattering experiments [4]. Besides its polemic nature, ZP ordering suggests reconsidering also the magnetic properties of charge ordered manganites [3,5].

Concerning the magnetic ground state, Zener polaron ordering should be reflected by *non collinear* arrangement of ferromagnetic entities. However, we found from previous neutron powder diffraction studies, that *collinear* and non *collinear* solutions are simply not distinguishable in $R_{½}Ca_{½}MnO_3$ [6]. Still, this distinction was possible in $YBaMn_2O_6$, were we surprisingly obtained a *non-collinear* arrangement of ferromagnetic square plaquettes of four Mn (Fig.1c) attributable to the presence of Zener polarons of four Mn in this compound [7].

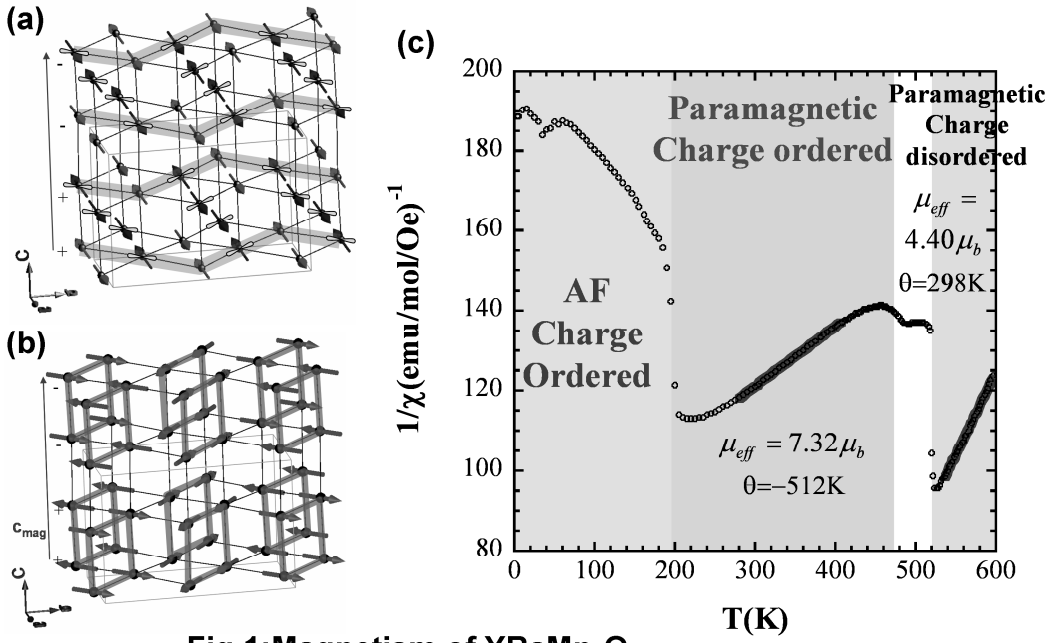

Fig.1:Magnetism of $YBaMn_2O_6$

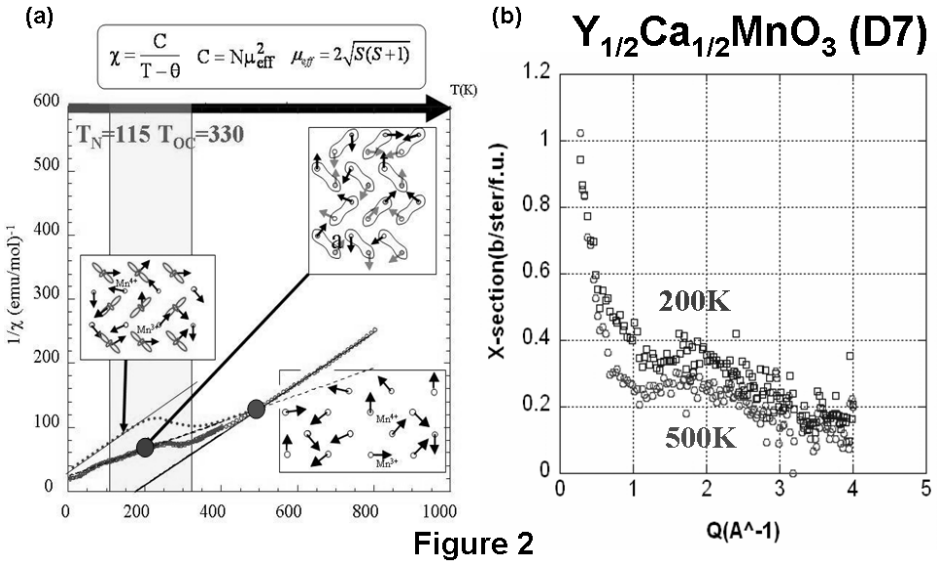

Figure 2

Besides, magnetic susceptibility measurements provide a more generic justification: both $Y_{1/2}Ca_{1/2}MnO_3$ [3] (Fig.2a) and $YBaMn_2O_6$ [6] (Fig2c), shows increases of the effective paramagnetic moments below the CO transition fitting the building up of large super-paramagnetic units of 2 (S=7/2) and 4 (S=7) ferromagnetically coupled Mn respectively.

A preliminary study of the change of magnetic correlations in $Y_{1/2}Ca_{1/2}MnO_3$ has been attempted from test beam time on D7, by extracting the pure magnetic scattering cross section at two temperatures across the charge ordering transition with the XZY polarization analysis. The data (Fig.2b) definitively manifests qualitative changes especially at low Q's, which indicate a change of form factor due to the delocalization of spin polarized electrons on the ferromagnetic unit. However, the effect is convoluted with the change of magnetic correlations.

Proposed experiment

In the light of the above, we propose first a straightforward qualitative comparison of the previous D7 data with data that we would collect on $YBaMn_2O_6$. We aim to evidence by this, possible differences associated to the role that can play the building of ferromagnetic entities of different sizes in different charge ordered manganites. This is required to judge the feasibility of doing a proper theoretical modeling of the data. Ultimately, we intend to use Monte-carlo simulation to compare the two compounds, and distinguish between realistic models for magnetic exchange using high spin Mn ions and ferromagnetic units respectively.

Using a furnace (T>300K), we need 3-4 temperatures per sample with about a day of measurement per temperature. In order to get extract the pure magnetic scattering signal on $YBaMn_2O_6$ and complete our preliminary measurements on $Y_{1/2}Ca_{1/2}MnO_3$ we ask for 7 days of beam time on D7.

References

[1] J. B. Goodenough, Phys. Rev. **100**, 564 (1955).
[2] T. Arima *et al*., Phys. Rev. B **66**, 140408 (2002)
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[4] S. Grenier *et al.* Phys. Rev. B **69**, 134419 (2004)
[5] G. Zheng et al., Phys. Rev. B **67**, 220404 (2003)
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[7] A. Daoud-Aladine *et al.* (unpublished)

Publication

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Proposal: **5-53-130** **Council:** 10/2006

Title: Change of magnetic correlations across charge ordering transtions in manganites from polarized neutron powder diffraction

This proposal is a new proposal

Researh Area: Physics

Industry: **Not related to industrial application**

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New neutron user? No
New ILL user? No

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Instrument	Req. Days
D7	7

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Zaharko O, Ronnow H, Mesot J, Crowe SJ, Paul DM, Brown PJ, Daoud-Aladine A, Meents A, Wagner A, Prester M, Berger H, PHYSICAL REVIEW B 73 (6): Art. No. 064422 FEB 2006

Zaharko O, Daoud-Aladine A, Streule S, et al.
PHYSICAL REVIEW LETTERS 93 (21): Art. No. 217206 NOV 19 2004

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