Investigation of Spin Dynamics in the *E-type* Antiferromagnetic Ground State of Mn-doped $Sr_3Ru_2O_7$

The aim of this proposal is to perform an inelastic neutron scattering (INS) investigation of $Sr_3(Ru_{1-x}Mn_x)_2O_7$ (x = 0.16)) single crystal using 7 days of beam time at the BT-7 Double Focusing Triple-Axis Spectrometer T7 in order to characterize the spin dynamics of this system. We have already completed an extensive elastic neutron scattering study for this doping level by using the awarded beam time [the result has been published in Phys. Rev B, Rapid Comm. **85**, 180410(R) (2012)]. Our investigation revealed the ground state of this system to have a Mn-induced unusual spin arrangement: *single-bilayer E-type antiferromagnetic (AFM) order*---a quasi 2D spin structure. However, the AFM order parameter, i.e. the staggered magnetization, cannot be described by the standard 2D magnetic phase transition picture. Understanding of the anisotropic spin-spin couplings and unusual electronic properties in this unique system can only be achieved by a comprehensive study of spin dynamics. Some preliminary INS results at measured at BT7 before shows unusual spin dynamics in this quasi-2D AFM ground state.

Scientific Background

The material reported in this investigation belongs to the much-studied Ruddlesden-Popper (RP) series $Sr_{n+1}Ru_nO_{3n+1}$, where n is an integer [1]. The RP series displays an abundant array of physical phenomena (as n changes) such as metallicity, spin-orbital ordering, and exotic superconductivity, all of which have re-fueled the interest in these and related ruthenates among the scientific community. To illustrate this, we have for example, the (n=*1*) member of the series and parent compound Sr_2RuO_4, which displays characteristics of both a paramagnetic Fermi liquid and of an unconventional spin-triplet superconductor (possibly linked to ferro- or antiferromagnetism) below 1.5 K [2]. The other end member of the series, namely the (n=∞) $SrRuO_3$, is an itinerant ferromagnet with a Curie temperature (T_C) of 160 K [3].

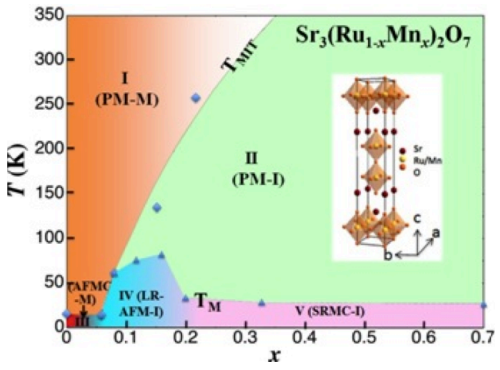

FIG. 1. Phase diagram of $Sr_3(Ru_{1-x}Mn_x)_2O_7$ from transport measurements, where T_{MIT} marks the metal-insulator transition and T_M the magnetic phase transition. PM-M stands for paramagnetic metal, PM-I paramagnetic insulator, AFM-I antiferromagnetic insulator and AFMC-M antiferromagnetically correlated metal. The inset represents the crystallographic structure of the system in the parent compound.

Between the two previously mentioned ruthenates lies the bilayer perovskite (n=*2*) $Sr_3Ru_2O_7$ (SRO237). This system is a paramagnetic metal with strong FM fluctuations in its ground state and it's characterized by Fermi-liquid behavior for temperatures bellow 10 K [4]. Recently, in a study by S. A. Grigera *et al* [5], $Sr_3Ru_2O_7$ was reported to exhibit a new quantum critical behavior—referred to as quantum critical end point (QCEP)—related to a metamagnetic (i.e. magnetic field-tuned) transition. Associated with this metamagnetic transition (and induced from a further decrease in temperature) a new phase—an electronic-like nematic phase—has been observed [5,6]. In this regime, thermal conductivity [7] and quantum oscillation [8] measurements suggest that ferromagnetic (FM) fluctuations are responsible for the non-Fermi liquid behavior expected at the QCEP. However, neutron diffraction measurements [9] have found no evidence of either long/short-range AFM or FM ordering in the range of temperatures from 1.4 to 125 K and reported an upper limit of 0.05 μ_B/Ru atom for any possible (if

any) ordered moment in the SRO 327 system. In contrast to the latter, two independent inelastic neutron scattering studies on the same parent compound revealed incommensurate magnetic fluctuations attributed to strong Fermi surface nesting [10, 11]. The above leads us to conclude, that an understanding of the magnetism even in the parent compound and specially that of the nature of the relevant magnetic fluctuations that drive the system into the QCEP in this regime remains elusive.

The magnetic fluctuations present in the system can be stabilized by doping with a *3d* transition-metal atom (Mn) on the Ru site in $Sr_3Ru_2O_7$ (Ru^{4+}, $4d^4$) [12]. Extensive transport and magnetization measurements have led to the phase diagram reported in figure 1 **[**14]. Our elastic neutron scattering experimental results [15] indicate that the effect of Mn-doping in this layered system is rather intriguing. We have found that, for $Sr_3(Ru_{1-x}Mn_x)O_7$ with $x = 0.16$, the magnetic structure in the ground state is that of an *E-type* AFM spin ordering with magnetic moments aligned along the c-axis [see **Fig 2**] and a Néel Temperature (T_N) = 82 K ,which is substantially lower than the reported metal-insulator transition temperature (T_{MIT}) = 140 K obtained from transport measurement [14]. The AF order is found to exhibit long-range order along the *ab* plane with an average ordered moment of $0.70\mu_B$/Ru site. On the other hand, the AFM correlation is only single bilayer-thickness along the *c* direction, which indicates quasi-2D behavior [15] (see **Fig. 3**). However, the AFM order parameter, i.e. the staggered magnetization, cannot be described by the standard 2D magnetic phase transition picture. To understand the nature of such unusual spin structure and magnetic properties, determining the spin-spin exchanging coupling strength along different high-symmetry directions and possible coupling with other degrees of freedom (such as lattice) is essential. Therefore, measurements on spin dynamics like spin waves in AFM phase, which has been conducted before, is the main focus of this proposal.

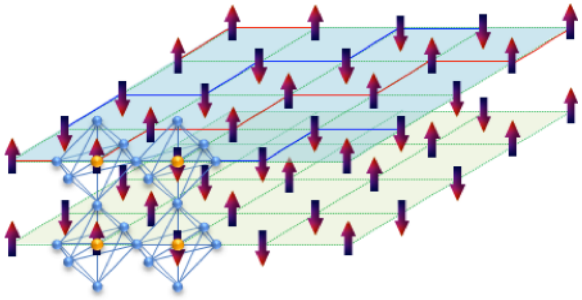

FIG. 2 (a) Schematic diagram revealing the spin texture of the $Sr_3(Ru_{0.84}Mn_{0.16})_2O_7$ sample. The magnetic unit cell is delimited by the octahedral (more than one unit cell is shown in order to make evident the zigzag chains of the *E-type* AFM structure).

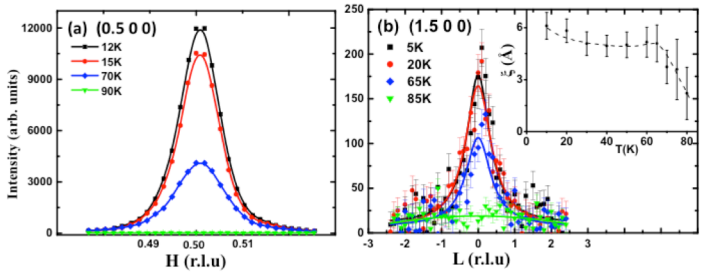

FIG. 3 Neutron diffraction profiles at magnetic Bragg peaks (a) (0.5, 0, 0) in (H,K,0) scattering plane and (b) (1.5, 0, 0) in (H,0,L) for various temperatures. The narrow (resolution limited) linewidth of the H scans across the (0.5, 0, 0) peak at low temperatures is indicative of long-range AFM ordering in the basal plane. The L scans across the (1.5, 0, 0) magnetic peak on the other hand exhibit very broad Lorentzian-profiles; the extracted c-axis magnetic correlation length $\xi(T)$ ploted in Fig. 3(d) inset shows only a maximum value of $\xi \sim 5$-6 Å bellow T_N.

Preliminary INS results

From our preliminary measurements with inelastic neutron scattering, we have observed well defined spin-wave excitation near the zone center. In addition, a sizable spin gap (~ 4 meV) is also observed, indicating large anisotropy in spin-spin interaction, consistent with observed quasi-2D static spin structure in the AFM ground state.

Detailed Description of the Proposed Experiment

We propose to use the BT-7 Triple Axis Spectrometer to continue our magnetic excitation study. The high flux from double focusing monochromator of the instrument is particular beneficial since the moment in the ordered state is relative small (~0.7 μ_B). We request 7 days to explore the complete spin wave dispersion relations along high symmetry directions and its temperature dependent behavior. The measurements include the dispersion, linewidth, and their temperature-dependence. We would like to address the following questions: 1) Is the magnetic Hamiltonian well characterized by local Heisenberg model? 2) Is there any magnetic frustrations giving rise to the unusual E-type magnetic configuration? 3) What is the strength of the exchange interactions within the bi-layered block? 4) Is the magnetic exchange interaction between layers negligible so the system can to be regarded as 2D? The dispersion results will yield values for the spin wave stiffness constant and of the exchange interaction as a function of direction, which we will then compare to those present in other quasi- 2D systems. The spectra weight, especially the q-dependence of linewidth, allows revealing the spin-spin correlations and possible coupling to other active degrees of freedom. One exciting issue is the Mn dopant effect on magnetic interaction [16,17].

References

[1] S.N. Ruddlesden and P. Popper, *Aca Crystallogr.* **11,** 54 (1998)
[2] K. Ishida *et al., Nature* **396,** 658 (1998) and references therein; Y. Sidis *et al., Phys. Rev. Lett.* **83,** 3320 (1999); I. I. Mazin and D. J. Singh, *PRL* **82,** 4324 (1999); Y. Maeno *et al., Nature (London)* **372,** 532 (1994); A. P. Mackenzie and Y. Maeno, *Rev. Mod. Phys* **75,** 657 (2003)
[3] A. P. Mackenzie *et al., Phys. Rev.* B **58,** R13318 (1998); P.B. Allen *et al., ibid* **53,** 4393 (1996)
[4] S.I Ikeda *et al., Phys. Rev.* B **62,** R6089 (2000)
[5] S. A. Grigera *et al., Science* **294,** 329 (2001); C. Pfleiderer *et al., Nature* **414,** 427 (2001); B. Binz and M. Sigrist, *Europhys. Lett.,* **65,** 816 (2004)
[6] R.A. Borzi *et al., Science* **315,** 214 (2007)
[7] F. Ronning *et al., Phys. Rev. Lett.* **97,** 067005 (2006)
[8] R.A. Borzi *et al., Phys. Rev. Lett.* **92,** 216403 (2004)
[9] Q. Huang *et. al., Phys. Rev* B **58**, 8515 (1998) and references therein.
[10] M.B. Stone *et al., Phys. Rev.* B **73,** 174426 (2006)
[11] L. Capogna *et al., Phys. Rev.* B **67,** 012504 (2003)
[12] Mathieu *et al., Phys. Rev.* B **72,** 092404 (2005)
[13] M. A. Hossain *et al., Phys. Rev. Lett* **101,** 016404 (2008)
[14] Biao Hu *et al., Phys. Rev.* B **81,** 184104 (2010) and references therein;
Biao Hu *et al., Phys. Rev.* B **84**, 174411 (2011)
[15] Dalgis Mesa, Feng Ye, Songxue Chi, J. A. Fernandez-Baca, W. Tian, Biao Hu, R. Jin, E. W. Plummer, and Jiandi Zhang, *Phys. Rev* B, Rapid Comm. **85**, 180410(R) (2012).
[16] Guorong Li, Qing Li, Minghu Pan, Biao Hu, Chen Chen, Jing Teng, Zhenyu Diao, Jiandi Zhang, Rongying Jin and E.W. Plummer, *Nature Scientific Report* **3**, 2882 (2013)
[17]M.A. Hossain et al., *Nature Scientific Report* **3**, 2299 (2013)