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# Magnetic field dependent neutron powder diffraction studies of $Ru_{0.9}Sr_2YCu_{2.1}O_{7.9}$

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Temperature and magnetic field dependent neutron diffraction has been used to study the magnetic order in $Ru_{0.9}Sr_2YCu_{2.1}O_{7.9}$. The appearance of (1/2, 1/2, 1/2), (1/2, 1/2, 3/2), and (1/2, 1/2, 5/2) peaks below $T_M=140$ K manifests the antiferromagnetic order. Neutron diffraction patterns measured in applied magnetic fields from 0 to 6 T show the destruction of the antiferromagnetic order with increasing field. There is no evidence of spontaneous or field-induced long range ferromagnetic order. This latter result contradicts the vast majority of other experimental observations for this system. © *2010 American Institute of Physics*. [doi:10.1063/1.3365576]

The high temperature Ru-based superconductors, $RuSr_2RCu_2O_8$ (Ru1212) (Refs. 1–3) and $RuSr_2R_{2-x}Ce_xCu_2O_{10}$ (Ru1222),[6–8] where R=Gd, Eu, Sm, and Y are also called superconducting ferromagnets due to the coexistence of superconducting and ferromagnetic (FM) order. Ru1212 exhibits magnetic ordering around $T_M=133-140$ K, and a superconducting transition takes place at $T_c=15-50$ K depending on the composition and preparation conditions.[1–3] There have been a number of conflicting reports concerning the magnetic order in Ru1212. The magnetization versus applied field curves of Ru1212 exhibit hysteresis below $T_M$, indicating FM order.[4,5] A zero-field muon spin-rotation study[3] reported low-field FM order in the ab plane. On the contrary, powder neutron diffraction measurements performed by Lynn *et al.*[1] on Ru1212Gd revealed low-field AFM order (G-type AFM) in the $RuO_2$ layers with the Ru moments aligned along the c-axis, and magnetic moment per Ru ion was estimated to be $1.18\mu_B$.[1]

In this work, temperature and magnetic field dependent neutron powder diffraction (NPD) studies have been carried out on $Ru_{0.9}Sr_2YCu_{2.1}O_{7.9}$ (Ru1212Y) to determine the magnetic structure and to investigate the magnetic response to external magnetic fields. To the best of our knowledge, the only report of field dependent neutron diffraction on $RuSr_2GdCu_2O_8$ was by Lynn *et al.*[1]

A polycrystalline sample of Ru1212Y was prepared by solid-state reaction using high pressure (~6 GPa) and high temperature (~1450 K) technique at National Institute for Material Science (NIMS), Japan.[9,10] This is the same sample used in previous magnetic susceptibility and neutron diffraction studies.[10,11] The sample has an onset superconducting transition $T_c$ of 40 K and magnetic ordering temperature $T_M$ of 140 K.[9,10]

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The present neutron data of ~1.58 g of the powder Ru1212Y sample were collected at two different neutron facilities, viz., the focusing diffractometer (E6) at the Berlin Neutron Scattering Center (BENSC) at Helmholtz-Zentrum-Berlin using wavelength λ=2.45 Å and the high intensity powder diffractometer (*Wombat*) installed at the *Opal* research reactor at Australian Nuclear Science and Technology Organization (ANSTO) with the incident neutron wavelength of 2.41 Å. Neutron diffraction pattern of Ru1212Y was collected by *Wombat* from $2\theta=16°$ to 135° in the temperature range of 20–160 K with 1 K step size. Figure 1 shows a portion of the diffraction pattern. In addition to the nuclear Bragg peaks indexed as (0 0 2), (0 0 3), and (1 1 0), additional peaks appear below $T_M=140$ K at $2\theta=27.5°$, 33°, and 41.5°. These peaks can be indexed as (1/2, 1/2, 1/2), (1/2, 1/2, 3/2), and (1/2, 1/2, 5/2), respectively. The appearance of additional peaks along with the Bragg peaks suggests antiferromagnetic (AFM) order with doubling of the unit cell in all directions. The comparison of present neutron data of

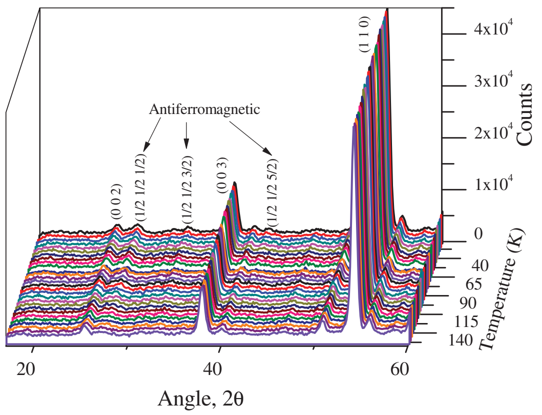


FIG. 1. (Color online) Neutron diffraction pattern of $Ru_{0.9}Sr_2YCu_{2.1}O_{7.9}$ sample in the temperature range of 20–160 K collected at ANSTO.

 

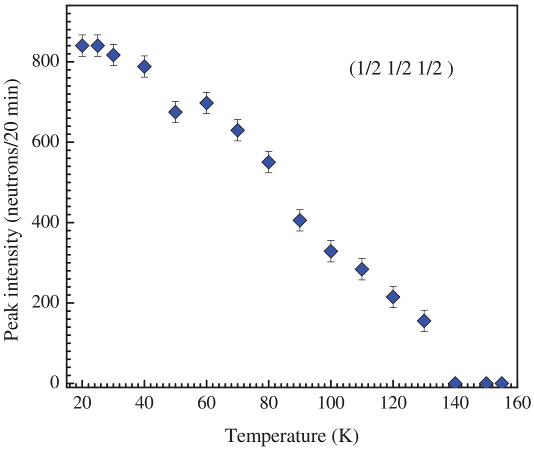


FIG. 2. (Color online) Intensity of AFM (1/2, 1/2, 1/2) peak as a function of temperature at zero field.

Ru1212Y with the neutron data reported by other groups on the same compound shows that our neutron data exhibit (1/2, 1/2, 1/2), (1/2, 1/2, 3/2), and (1/2, 1/2, 5/2), AFM peaks, whereas only a (1/2, 1/2, 1/2) peak was observed by Takagiwa *et al.*,[12] and (1/2, 1/2, 1/2) and (1/2, 1/2, 3/2) magnetic peaks were reported by Yelon *et al.*[11] This is further in agreement with the neutron diffraction data reported by Lynn *et al.*[1] on $RuSr_2GdCu_2O_{8-\delta}$, which showed (1/2, 1/2, 1/2) and (1/2, 1/2, 3/2) reflections at the Ru ordering temperature of about 135 K. Out of these three additional peaks, the (1/2, 1/2, 1/2) peak is the strongest; hence this peak will be used for further discussion. In order to study the behavior of (1/2, 1/2, 1/2) peak with changing temperature, its intensity was plotted as a function of temperature in Fig. 2. Below $T_M = 140$ K, the intensity of AFM peak increases with decreasing temperature. Contrary to this evidence of AFM order, M(H) loops of Ru1212Y exhibit FM-like hysteresis loops;[11] therefore, it is important to search for the FM order in the system. In neutron diffraction, FM is manifested as additional scattering at nuclear Bragg peaks. No change was observed in the intensity of nuclear Bragg peaks, where a FM component would appear; hence no indication of FM order was given by NPD measurements. Similar observation was made by Lynn *et al.*, and their results gave an upper limit of $0.1\mu_B$ to any FM component. Such a small value of FM moment may remain undetected by NPD.

Although the temperature dependent NPD patterns of Ru1212Y have been reported previously,[10,11] no field dependent NPD study has been published. The only study of this type was reported on $RuSr_2GdCu_2O_{8-\delta}$ by Lynn *et al.*[1] In order to investigate how the Ru1212Y system behaves in the presence of external magnetic field in the superconducting state, NPD patterns were measured far below the superconducting and magnetic order temperatures of Ru1212Y. Neutron data were collected at $T = 1.5$ K under the applied field of 0–5 T at BENSC. The measurement at each field took approximately 3 h. Figure 3 shows NPD pattern at $T = 1.5$ K for $H = 0$, 1, 2, and 3 T. It is clear that the magnitude of the AFM peaks (1/2, 1/2, 1/2) and (1/2, 1/2, 3/2) reduces with increasing field. In order to deduce the effect of external magnetic field on the nuclear Bragg peaks, the NPD data

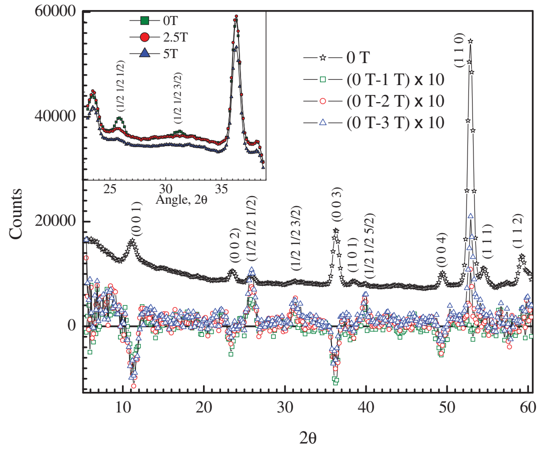


FIG. 3. (Color online) Subtraction of neutron diffraction patterns of 1, 2, and 3 T, respectively, from 0 T neutron diffraction pattern of $Ru_{0.9}Sr_2YCu_{2.1}O_{7.9}$ sample collected at BENSC. Inset shows neutron diffraction pattern between $2\theta = 22°$ and 45° measured at 1.5 K for applied fields of 0, 2.5, and 5 T.

collected at 1, 2, and 3 T were subtracted from 0 T NPD data and plotted in Fig. 3. The difference patterns exhibit positive peaks corresponding to the AFM reflections, indicating that the peak intensity decreases when the magnetic field is applied. With increasing field there is a nonlinear decrease in the AFM peaks, which may indicate a diminishing AFM order. Previously, NPD patterns of Ru1212Gd measured by Lynn *et al.*[1] in the fields ranging from 0 to 7 T showed similar results. Figure 3 also shows that the external magnetic field increases the intensity of the (0 0 1), (0 0 2), (0 0 3), and (0 0 4) nuclear peaks. For the Ru1212Gd sample, Lynn *et al.*[1] could observe additional scattering only at (0 0 2) nuclear peak when external field was applied. The additional scattering of (0 0 *l*) nuclear peaks in an applied magnetic field may indicate a field-induced FM. Since only the component of magnetic moments perpendicular to the scattering vector can be detected by NPD, the induced ferromagnetism would be perpendicular to the c-axis (consistent with the proposition of Bernhard *et al.*[3]).

However, we also note that the (1 1 0) nuclear reflection decreases strongly with increasing field. Given that there is no FM order in zero field and that the AFM order is G-type, the only possible interpretation of this is that the field induces grain reorientation with c-axis aligned perpendicular to the magnetic field. The evidence of grain reorientation brings into question whether the application of a magnetic field really induces the disappearance of AFM order or the appearance of FM order.

We also note that there was an overall reduction in the background of the diffraction pattern in applied field of 5 T (see Fig. 3 inset). This indicates that the sample was highly magnetized and levitating inside the sample holder partly lifted out the neutron beam. Returning to zero field, all the peaks recover their zero-field intensities, indicating that the effect of the field is reversible and also that no preferred orientation of the loose powder particles occurred when the field was applied. NPD measurements of Ru1212Gd conducted by Lynn *et al.* did not report any shift in the back-

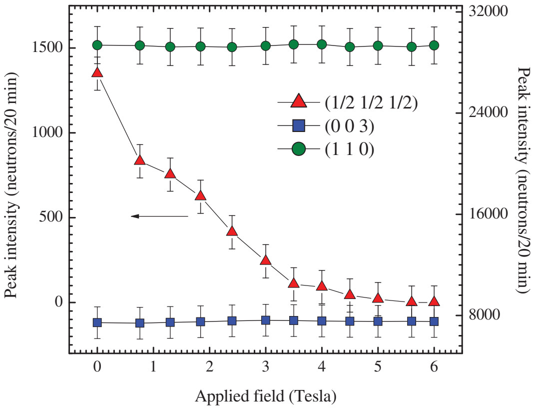


FIG. 4. (Color online) Intensity of (1/2, 1/2, 1/2), AFM peak and (0 0 3) and (1 1 0) nuclear Bragg peaks as a function of applied field, as measured on a tightly packed sample.

ground of the high field data. Their results showed that with an increase in field, the intensity of the AFM reflection begins to decrease, while the induced magnetization increases. When the field was increased from 0 to 7 T, there is no significant AFM intensity observed, but there was induced magnetization with a net moment of $1.4\mu_B$ perpendicular to c-axis. Authors proposed that the Ru moments rotate into another AFM (spin-flop) structure rather than becoming fully aligned with the field. The NPD study of Ru1212Y is consistent with the NPD study of Ru1212Gd sample, except the fact that the Ru1212Y sample gets highly magnetized for fields as high as 5 T. This may be because the paramagnetic moments of Gd may not allow Ru1212Gd sample to magnetize fully at high fields.

In order to extract meaningful and reliable information about magnetism in Ru1212 from neutron diffraction, we repeated the study of magnetic field dependence on the *Wombat* diffractometer, with a sample clamped tightly inside the vanadium sample holder by a cylindrical aluminum insert. Neutron data were collected at 10 K in the field ranging from 0 to 6 T. No shift in the background was observed for high field neutron diffraction data, which indicated that no grain levitation took place. The response of the (1/2, 1/2, 1/2), (0 0 3) and (1 1 0) diffraction peaks to applied field is plotted in Fig. 4. The intensity of (1 1 0) peak is clearly constant in the entire field range, confirming that no grain reorientation took place. The field dependence of AFM order [indicated by the drop in the intensity of (1/2, 1/2, 1/2) peak (Fig. 4)] shows almost complete destruction of AFM from 0 to 3.5 T. Further, Fig. 4 shows that the intensity of (0 0 3) nuclear Bragg peaks remains unaffected over the entire range of applied magnetic field, neither was any additional intensity detected in other Bragg peaks. This result confirms our conclusion that the neutron diffraction measurements carried out at BENSC were dominated by grain reorientation and levitation. Zero field, temperature dependent neutron diffraction studies of $Ru_{0.9}Sr_2YCu_{2.1}O_{7.9}$ sample showed that the system exhibits an AFM order below $T_M = 140$ K, but despite the evidence of FM from magnetization measurements, FM order could not be detected by neutron diffraction. In this case diffuse neutron scattering would be a vital tool to detect short range FM in the system. The field dependent neutron studies showed that the AFM order disappears with increasing field. The apparent field induced FM of the loosely packed Ru1212 powder was shown, by the study of the tightly packed powder, to be just grain reorientation, with moments perpendicular to c-axis.

In conclusion, our field dependent NPD measurement results further illustrate a puzzling response to magnetic fields of this system, which exhibits the hysteretic behavior observed in bulk magnetic measurements, but it seems not to manifest in long range FM ordering. Indeed, the vast majority of global and *in situ* probing techniques consistently exhibits the existence of the FM signal, whereas only neutron diffraction measurements robustly indicate AFM ordering. Clearly, the origin of this controversy is yet quite unclear and requires further investigation.

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[1] J. W. Lynn, B. Keimer, C. Ulrich, C. Bernhard, and J. L. Tallon, Phys. Rev. B **61**, R14964 (2000).
[2] A. C. McLaughlin, W. Zhou, J. P. Attfield, A. N. Fitch, and J. L. Tallon, Phys. Rev. B **60**, 7512 (1999).
[3] C. Bernhard, J. L. Tallon, Ch. Niedermayer, Th. Blasius, A. Golnik, E. Brcher, R. K. Kremer, D. R. Noakes, C. E. Stronach, and E. J. Ansaldo, Phys. Rev. B **59**, 14099 (1999).
[4] G. V. M. Williams and S. Krämer, Phys. Rev. B **62**, 4132 (2000).
[5] J. L. Tallon, J. W. Loram, G. V. M. Williams, and C. Bernhard, Phys. Rev. B **61**, R6471 (2000).
[6] I. Felner, U. Asaf, Y. Levi, and O. Millo, Phys. Rev. B **55**, R3374 (1997).
[7] R. Nigam, A. V. Pan, and S. X. Dou, Phys. Rev. B **77**, 134509 (2008).
[8] R. Nigam, A. V. Pan, and S. X. Dou, J. Appl. Phys. **101**, 09G109 (2007).
[9] V. P. S. Awana and E. Takayama-Muromachii, Physica C **390**, 101 (2003).
[10] H. A. Blackstead, W. B. Yelon, M. Kornecki, M. P. Smylie, Q. Cai, J. Lamsal, V. P. S. Awana, S. Balamurugan, and E. Takayama-Muromachi, Phys. Rev. B **76**, 094507 (2007).
[11] W. B. Yelon, Q. Cai, J. Lamsal, H. A. Blackstead, M. Kornecki, V. P. S. Awana, H. Kishan, S. Balamurugan, and E. Takayama-Muromachi, J. Appl. Phys. **101**, 09G104 (2007).
[12] H. Takagiwa, J. Akimitsu, H. Kawano-Furukawa, and H. Yoshizawa, J. Phys. Soc. Jpn. **70**, 333 (2001).