The Open Superconductor Journal, 2009, *1*, 25-32

Magnetic-Properties of $R\mathrm{Sr_2NbCu_2O_8}$; R = Nd, and Tb Compounds Prepared under Extreme Conditions

S. Balamurugan[*,#]

Advanced Nano Materials Laboratory, National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan

Abstract: The *niobio-cuprates* with starting composition, $R\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$; R = Nd, and Tb have been prepared under extreme synthesis conditions and characterized by powder X-ray diffraction technique and their magnetic properties were investigated briefly in this article. The present high-pressure synthesized $Nd\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$ (*Nd*/1212Nb) compound has been compared briefly with the reported ambient pressure prepared compound. X-ray diffraction patterns of $R\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$; R = Nd, and Tb compounds taken at room temperature clearly indicate that the space group *P*4/*mmm* (123) is likely for the structure. Magnetic susceptibility, (χ vs T) measurements on $Nd\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$ compound reveal likely (Pauli) paramagnetic-nature where as the magnetization versus field, M(H) data taken at low temperature (1.8 K) reveals non-linear increase of magnetization with respect to the applied magnetic field indicating possibly due to canted antiferromagnetic interaction in this Nd compound and a magnetic saturation is not seen, signifying no magnetic orders within the range. Although a comparable M(H) isotherm has been observed at selected temperatures for most of the high-pressure synthesized niobio-cuprates, they show different results on magnetic susceptibilities, specific heat and transport measurements. Interestingly, unlike $Nd\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$ compound, the $Tb\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$ (*Tb*/1212Nb) compound (known for the first time) specifies two antiferromagnetic-like transitions (T_N) at ~41 K as well as at ~ 8 K with branching of *zero-field-cooled (zfc)* and *field-cooled (fc)* magnetization and a *zfc* cusp like down turn in magnetization at ~8 K, followed by negative susceptibility at 2 K. The M(H) plot measured at 1.8 K and ±70 kOe conditions reveals abnormal (spin-flop) magnetization isotherm for *Tb*/1212Nb compound. The paramagnetic *Weiss* temperature, θ_w and the paramagnetic effective moment, p_{eff} obtained from the linear region of χ^{-1}(T) plots are: –64.07 K and 3.71 μ_B/Nd for Nd compound and –15.67 K and 9.42 μ_B/Tb for Tb compound, respectively. The negative sign of θ_w ascertains the antiferromagnetic correlations in these studied compounds.

Keywords: High-pressure-synthesis, *niobio*-cuprate, $R\mathrm{Sr_2NbCu_2O_8}$, physical-properties.

1. INTRODUCTION

Although the Sr-based 1212–type *niobio-cuprates* $\mathrm{NbSr_2}R\mathrm{Cu_2O_{8-\delta}}$, R = rare earth elements have been known in the literature since 1992, unlike the 1212 type *rutheno-cuprates* ($\mathrm{RuSr_2}R\mathrm{Cu_2O_{8-\delta}}$), the niobio-cuprates have not been studied to a large extent. There are few reports in 1212–type niobio-cuprates, $R\mathrm{Cu_2NbSr_2O_8}$; R = Nd, Pr, Gd, Eu, Sm compounds which are prepared *under normal pressure conditions* [1–8]. For the present investigation two different rare earth elements such as, *Nd* and *Tb* has taken up for the syntheses of $R\mathrm{Cu_2NbSr_2O_8}$; R = Nd (*Nd*/1212Nb), and Tb (*Tb*/1212Nb) niobio-cuprate compounds under high-pressure. To the best of our knowledge there is no report in R = Tb compound whereas for R = Nd compound there are few reports in literature [1–4]. In this paper more emphasis is given in *Nd*/1212Nb compound than *Tb*/1212Nb. Though the former phase had known to be crystallized in different space group, the later lacks its quality in sample preparation for precise investigation. The R = Nd compound crystallized in tetragonal symmetry with different space group *viz*., *P4bm* (100) [1] and *I4/mcm* (140) [2] and *P4/mmm* (123) [1] *under ambient synthesis conditions* (see Table **1**). Zandbergen *et al*. [1] reported that the structure of $\mathrm{NbSr_2}Nd\mathrm{Cu_2O_8}$ compound consists of layer sequence Nd–$\mathrm{CuO_2}$–SrO–$\mathrm{NbO_2}$–SrO–$\mathrm{CuO_2}$ having a perovskite like structure, which is quite similar to that of well known $Y\mathrm{Ba_2Cu_3O_{7-\delta}}$ superconductor. The structure of this $Nd\mathrm{Sr_2NbCu_2O_8}$ compound has been identified with electron diffraction as well as with high resolution electron microscopy techniques and reveal that it

Table 1. Unit Cell Parameters for $Nd\mathrm{Sr_2NbCu_2O_8}$ Compounds Crystallized in Tetragonal Symmetry with Different Space Group *viz.*, *P4/mmm* (123), *P4bm* (100) and *I4/mcm* (140) Under Ambient Synthesis Conditions

Space Group	*a* (Å)	*c* (Å)	*V* (Å)^3	Ref.
P4/mmm (123)	3.888	11.664	176.3	[3]
P4bm (100)	5.49	11.66	351.43	[1]
I4/mcm (140)	5.5037	23.3356	707.49	[2]

---

*Address correspondence to this author at the Advanced Nano Materials Laboratory, National Institute for Materials Science (NIMS), 1-1 Namiki, Tsukuba, Ibaraki 305-0044, Japan; E-mails: asbalamurugan@yahoo.co.in, scandium.chemistry@gmail.com, sbala288@yahoo.co.in

#Present address: Institut für Anorganische und Analytische Chemie, University of Münster, Corrensstrasse 30, D-48149 Münster, Germany

1876-5378/09 2009 Bentham Open

has an $a_p\sqrt{2}$, $a_p\sqrt{2}$, *c* superstructure (a_p is the *a*-axis of a simple cubic perovskite) which leads to a space group *P4bm* (100) owing to the rotation of NbO_6 octahedra along the *c*-axis [1]. The superstructure features of *Nd*/1212Nb compound are indistinguishable to the higher homologue compound $NbSr_2(Nd_{1.5}Ce_{0.5})Cu_2O_{10}$ [9].

The attempt by Kopnin *et al.* [3] to induce superconductivity in *Nd*/1212Nb compound after annealing at $P(O_2) = 1$ and 100 bar conditions failed to detect diamagnetic signal in magnetic susceptibility measurements down to 12 K. In this article the structural and magnetization results of *R* = Nd and Tb, compounds are presented and the high-pressure synthesized *Nd*/1212Nb compound data are compared with the available literature. In addition, though *R*= Tb compound could not be obtained in single-phase form, but it is quite good enough for the preliminary investigations and the data are briefly presented.

2. EXPERIMENTAL

Based on our previous work on $Nb_{0.9}Sr_2YCu_{2.1}O_{7.9}$ [10], in the present study polycrystalline samples of nominal compositions $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ with *R* = Nd and Tb samples were prepared through a high pressure and high temperature solid-state reaction route. The starting materials used were high purity (> 3N) fine powders of commercial Nb_2O_5, CuO, Cu, Nd_2O_3, Tb_4O_7, and laboratory prepared SrO_2, and $SrCuO_2$. These appropriate chemicals were homogeneously mixed in an agate mortar with pestle for 30 minutes in open atmosphere. After that the homogeneous reaction mixtures (about 300-400 mg) were tightly filled in gold (platinum) capsules. The high pressure experiments were performed in a flat-belt type (KOBELCO) sophisticated apparatus. Pyrophilitye was used as a pressure transmitting medium and graphite heater/cylinder as a high temperature furnace. After mounting the high-pressure cell on the apparatus, the pressure was slowly increased to 6 GPa in 15 minutes. Then the temperature was increased to 1200 - 1450° C in 15-20 minutes and soaked for 30 -180 minutes in order to obtain good quality samples. After the reaction time, the power was switched off and the pressure was released slowly in 90 minutes. The high pressure products were mechanically cleaved and polished with sand paper. The sintered (polished) samples were used for characterization. More details about syntheses of SrO_2 and $SrCuO_2$ precursor can be seen in our previous article [11].

For sample characterizations, the nominal composition $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ with *R* = Nd and Tb samples were used for structural and magnetization studies. All the *as*-prepared high-pressure samples were characterized for structure identification by powder X–ray diffraction (XRD) on a diffractometer using the Ni–filtered *Cu* K_α radiation. The *temperature* and *field* dependence of magnetization data were collected for all samples between 2 and 350 K with applied magnetic fields up to ± 70 kOe using magnetic property measuring system (MPMS/Quantum design).

3. RESULTS AND DISCUSSION

The powder X–ray diffraction (PXRD) patterns of $RSr_2NbCu_2O_8$; *R* = Nd, and Tb compounds measured at room temperature conditions are depicted in Fig. (**1**). It is clearly seen from the XRD patterns that both compounds crystallized in tetragonal structure with space group *P*4/*mmm*. The *Tb*-based niobio-cuprates failed to obtain in phase pure form, while *R* = *Nd* compound obtained (nearly) in single phase materials. Attempt to syntheses phase-pure samples on these two compounds were failed under few different synthesizes conditions. Neither stoichiometric phase $RSr_2NbCu_2O_8$ nor Nb-poor and Cu-rich phase $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ could be obtained in phase pure form in the present study. The lattice parameters calculated with space group *P4/mmm* for the *Nd*- and *Tb*-based compounds (see Table **2**) are *a* = *b* = 3.889(3) Å, and *c* = 11.64(1) Å for *R* = Nd and *a* = *b* = 3.868(5) Å, and *c* = 11.60(1) Å, respectively. A variation noticed in the lattice parameters are due to the difference in *R* ions and agreement with the ionic radii of *R* ions. For *R* = Nd, all the peaks could be well matched to the space group of *P*4/*mmm* (123) except one weak reflection noticed at $2\theta = 31.88°$ which is most probably due to the $SrNbO_3$ phase. The unit cell parameters of present high-pressure synthesized *Nd* samples are close agreement with the reported values of Kopnin *et al.* (*a* = *b* = 3.888(3) Å, and *c* = 11.664(1) Å, *V* = 176.32(6) Å^3) for their ambient pressure prepared sample [3]. The lattice parameters obtained from space group *P4bm* (*a* = *b* = 5.49 Å, and *c* = 11.66 Å, *V* = 351.4 Å^3) by Zandbergen *et al.* [1] and from *I4/mcm* (*a* = *b* = 5.5037 Å, and *c* = 23.3565 Å, *V* = 707.49 Å^3) by Vybornov *et al.* [2] for the ambient pressure prepared *Nd* samples are different from present high-pressure sample as well as with Kopnin *et al*. report [3]. For *R* = Tb compound though majority of the peaks are well matched with the main phase (*Tb/*1212Nb), a few weak reflections are observed at 2θ = 25.14, 31.62, 32.14 and 44.48°. All these impurity reflections are marked with symbol * in Tb sample's XRD pattern. Except one peak seen at $2\theta = 32.14°$ is due to $Sr_2Nb_2O_7$ all other three impurity peaks belong to $SrCuO_2$. If we closely look the XRD pattern of *Tb*-based sample one can judge that the sample is almost good enough for the preliminary investigations. Except two impurity peaks (2θ = 31.62 and 32.14°), other two peaks intensity level is close to the background level. However the quality of the samples should be improved for *precise interpretations*. Varying the preparation (annealing treatment) under wide conditions as well as controlling the oxygen content under high-pressure may yield phase-pure *Tb*-based 1212-type samples. If we carefully look the XRD pattern of *Tb*-sample, one can notice that all the peaks are slightly broadened as compared to *Nd*-based sample. It seems that the present applied high-pressure (6 GPa) and high temperature (1450ºC) annealing treatment is not enough to produce phase-pure samples. The isostructural tetragonal *Tb*-based *irido-cuprate,* $IrSr_2TbCu_2O_8$ compound synthesized by Dos santos-Garcia *et al*. [12] under high pressure of 9.2 GPa did not yield the single phase material. They failed to obtain a phase pure sample and the main tetragonal phase contained with some impurity phases. In fact the sample quality was nearly same as to the present *Tb/*1212Nb compound prepared under 6 GPa high pressure. If we perform several high pressure experiments above 6 GPa like *Tb*-based *irido-cuprate*, we may obtain phase pure sample. This effort may bring definite conclusion for the exciting data analysis.

The zero field cooled (*zfc*) and field cooled (*fc)* curves in χ vs *T* measurements under H = 10 Oe field indicate that the

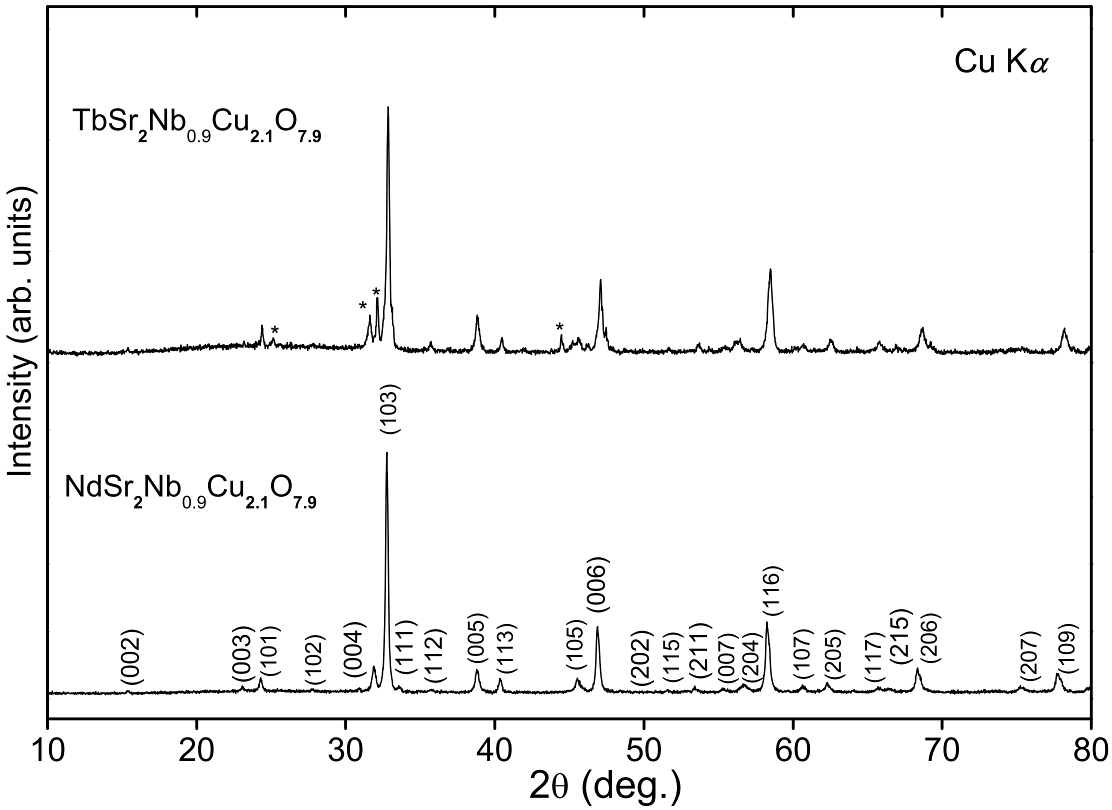

Fig. (1). Room temperature powder XRD patterns for $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$; R = Nd and Tb compounds prepared *under high-pressure medium*.

Table 2. Structural and Magnetic Parameters for $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$; R = Nd and Tb Compounds Prepared Under High-Pressure Medium

Compound	Unit Cell Parameters			Magnetic Parameters		
	a (Å)	*c* (Å)	*V* (Å)^3	θ_w (K)	p_{eff} (μ_B)	*M* ((μ_B/*f.u.*)
Cu_2*Nd*NbO_8Sr_2	3.889(3)	11.64(1)	176.0(9)	-64.0(7)	3.71	2.13
Cu_2*Tb*NbO_8Sr_2	3.868(5)	11.60(1)	173.6(1)	-15.6(7)	9.42	4.18

Nd/1212Nb compound is likely paramagnetic-in-nature (Pauli) with relatively very low susceptibility value in the order of 10^{-4} (see Fig. **2** main panel). It is also seen in the Fig. (**2**) main panel that both *zfc* and *fc* curves do not superimpose each other around 20 – 60 K temperature range in the χ vs T measurements and it indicates that the sample is not purely paramagnetic-in-nature. On the other hand, the $\chi(T)$ plot for the *Tb*-based *niobio-cuprate*, *Tb*$Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound exhibits that the compound is in a mixed magnetic state (see inset of Fig. **2**). It indicates two antiferromagnetic-like transitions ($T_{mag.}$) at ~41 K as well as at ~ 8 K with branching of *zfc* and *fc* magnetization and a *zfc* cusp like down turn in magnetization at ~8 K, followed by negative susceptibility around 2 K. The magnetic susceptibility in *zfc* measurement increases with decreasing the temperature from 300 K till ~ 100 K, below 100 K the raise in susceptibility with respect to temperature decrease is relatively high as compared to the high temperature region and reaches maximum of about 2.74 × 10^{-4} emu/g.Oe at 41 K then starts to decrease and undergoes a second antiferromagnetic transition (T_2) at ~ 8 K. Below this temperature a rapid decrease in magnetization signal is noticed in *zfc* curve and at the low temperature, the system reaches a negative susceptibility state. That is at 2 K the magnetic susceptibility signal is about –7.2 × 10^{-5} emu/g.Oe. In the *fc* mode the compound undergoes first antiferromagnetic (slightly broad) transition at ~ 41 K like *zfc* curve and decreases to 20 K. Below 20 K there is a sudden rise in the susceptibility signal and shows a second antiferromagtic transition at 5 K. This complex magnetic anomaly is different from *Nd*/1212Nb compound. Now a question arises from the interpretation of the magnetization data that whether it is really from the Tb/1212Nb phase or from any other secondary phase? We believe that it is due to Tb/1212Nb phase. But one cannot rule out other possibility also. Since it is a preliminary report in Nb1212Tb system, in order to strengthen the discussion on magnetism of the system we stress for further studies.

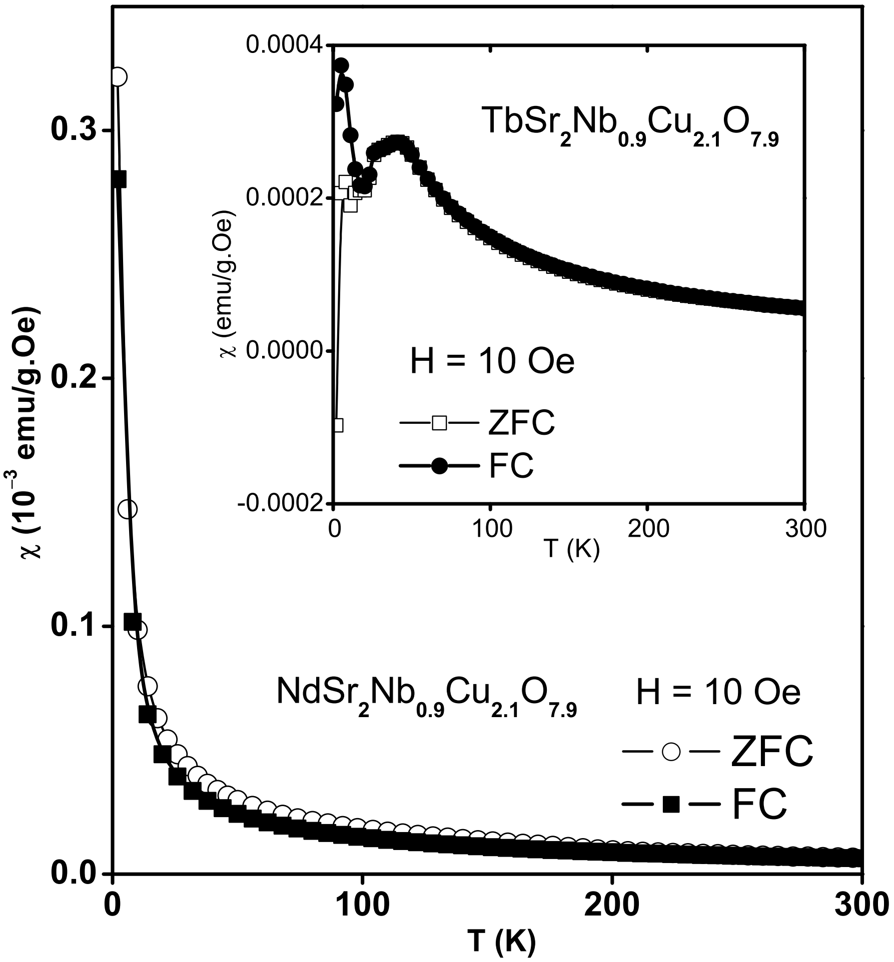

Fig. (2). Magnetic susceptibility vs. temperature (χ vs T) curves for $NdSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound measured in both FC and ZFC modes at 10 Oe. Inset shows same for the $TbSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound.

In order to evaluate the Weiss (paramagnetic) temperature as well as the effective paramagnetic moment for free Nd^{3+} and Tb^{3+} ions in the presently studied two compounds (R = Nd and Tb), the temperature dependence of molar magnetic susceptibility, $\chi(T)$ (left axis) and inverse molar magnetic susceptibility, $\chi^{-1}(T)$ (right axis) curves are depicted in Fig. (**3**). These measurements were taken up under the higher applied field of H = 10 kOe in both *zfc* and *fc* conditions between 2 and 350 K. For R = Nd, the $\chi(T)$ and $\chi^{-1}(T)$ data clearly show that there are no magnetic anomalies in both *zfc* and *fc* plots, whereas R = Tb compound shows antiferromagnetic transition at T_N = 5 K (see Fig. **3b** left axis) in both *zfc* and *fc* curves. It is noted that the antiferromagnetic (slightly broad) transition seen for R = Tb at T_N = 41 K in low field (H = 10 Oe) measurements disappeared under higher applied field, H = 10 kOe. Unlike the low field $\chi(T)$ measurements, no *zfc*-*fc* branching is seen in the high field H = 10 kOe $\chi(T)$ measurements down to 2 K. For the *Nd*-based sample the inverse molar susceptibility curve follows Curie-Weiss behavior above 50 K as depicted in the opposite to the $\chi(T)$ plot and yields the Weiss paramagnetic temperature, θ_w about –64.07 K. The negative sign of θ_w indicates that the magnetic interactions are antiferromagnetic in the presently studied *Nd*-based compound. The paramagnetic moment ($p_{eff} = \sqrt{8C}$) obtained from the linear region of χ^{-1}(T) curve is 3.71 μ_B/Nd, which is close to the value expected for free trivalent *Nd* ions [13]. The magnetic parameters determined from the modified *Curie-Weiss* equation, $\chi = \chi_0 + C/(T-\theta)$ in the temperature regime between 4 and 300 K by Vybornov *et al.* [2] for their ambient pressure synthesized *Nd*/1212Nb compound crystallized in *I4/mcm* (140) space group resulted: $\theta_w = -13$ K, p_{eff} = 3.1 μ_B/Nd and temperature independent susceptibility, $\chi_0 = 2.02 \times 10^{-3}$ emu/mol. Their reported p_{eff} (3.1 μ_B/Nd) value is quite smaller than the present high-pressure synthesized sample (3.71 μ_B/Nd), which (former) might have affected by the crystal field effect. For the higher homologous series, $NbSr_2(Nd_{1.5}Ce_{0.5})Cu_2O_{10}$ it has been reported by Goodwin *et al.* [14] that the modified *Curie-Weiss* fit yielded $\theta_w = -19.6$ K, p_{eff} = 2.97 μ_B/Nd and $\chi_0 = 1.64 \times 10^{-5}$ emu/mol. The p_{eff} (2.97 μ_B/Nd) of *Nd*/1222Nb system is comparable to the report of Vybornov *et al.* [2] for the ambient pressure synthesized *Nd*/1212Nb compound.

Like *Nd*-based compound, the *Tb*-based compound also follows the Curie-Weiss behavior (almost entire temperature range) above 10 K in the inverse molar susceptibility curve (Fig. 3b left axis). The experimental values of θ_w and p_{eff} obtained from the linear region of χ^{-1}(T) plot are: –15.67 K and 9.42 μ_B/Tb, respectively. The observed p_{eff} value is close to the theoretical value of 9.72 μ_B/Tb for a free Tb^{3+} ion [13]. The higher homologous series (in Tb system), $NbSr_2(Tb_{1.5}Ce_{0.5})Cu_2O_{10}$ crystallized in tetragonal symmetry with

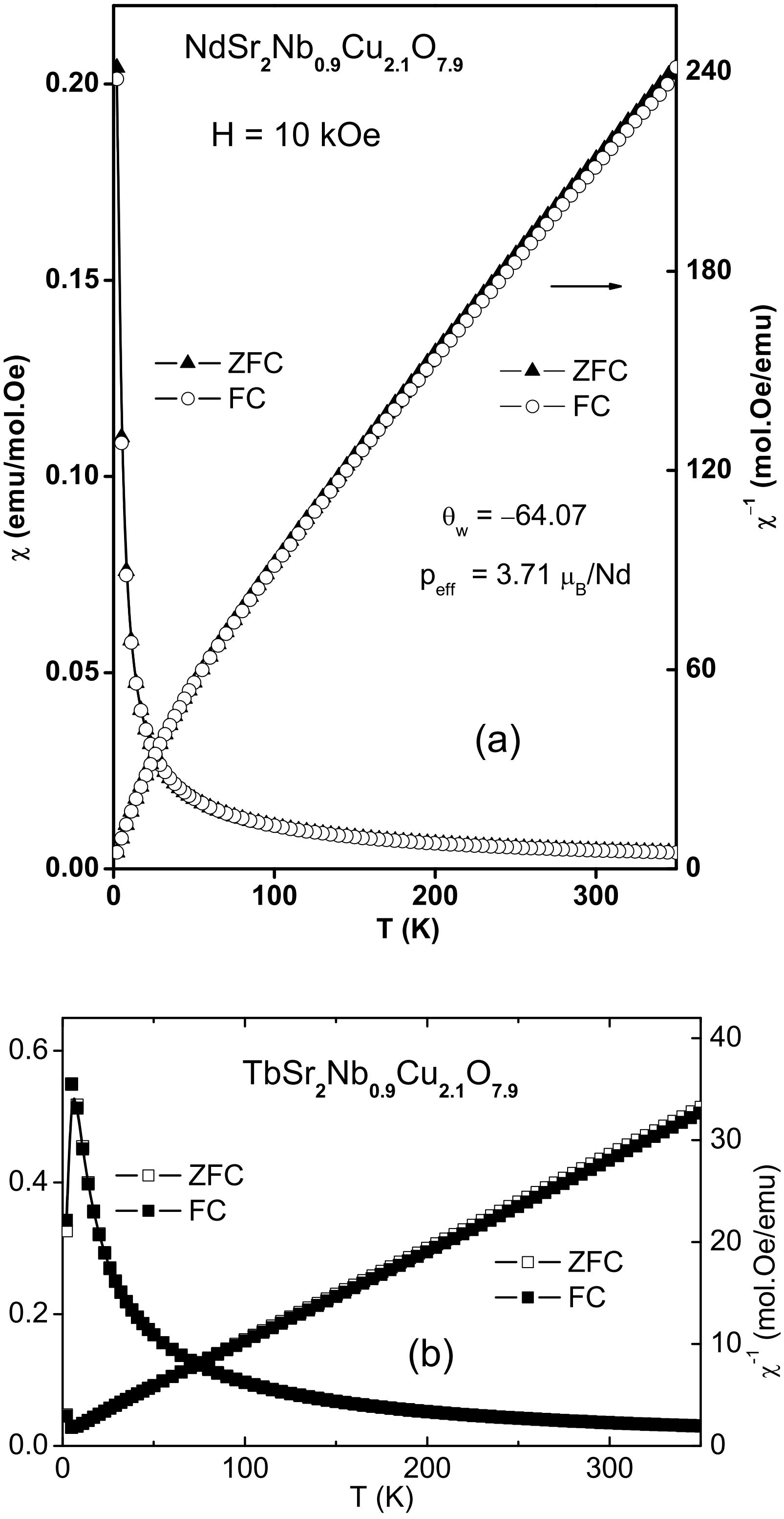

Fig. (3). The plots of molar magnetic susceptibility (left axis), χ and inverse molar magnetic susceptibility (right axis) χ^{-1} against temperature, (T) for the $R\mathrm{Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}}$: R = Nd **(a)** and Tb **(b)** compounds measured in both FC and ZFC conditions under the applied field of 10 kOe.

*I*4/*mmm* space group yielded θ_w is –6.64 K and p_{eff} = 9.74 μ_B/Tb. The negative sign of θ_w indicates that the magnetic interactions are antiferromagnetic.

In Fig. (**4**) the isothermal magnetization data *versus* field, M(*H*) curves taken for *R* = Nd compound at different temperatures such as, T = 1.8, 10, 30 and 50 K with higher

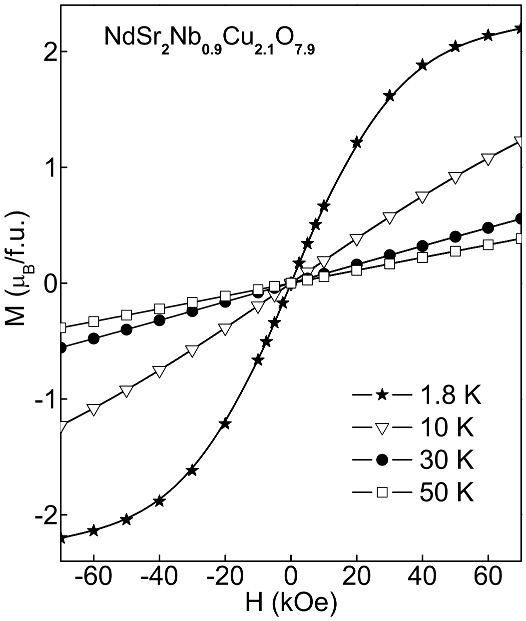

Fig. (4). Isothermal magnetization, M(H) curves measured at various temperatures in higher applied fields up to ± 70 kOe for *Nd*$Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound.

applied fields up to ±70 kOe; –70 kOe ≤ H ≤ 70 kOe are presented. Interestingly the M(H) curve measured at 1.8 K shows an increase of magnetization with increasing field up to 70 kOe with induced moments at the maximum magnetic field equal to 2.13 μ_B/f.u. indicating that the 70 kOe field is not enough to induce saturation of the magnetic moments in R = Nd compound. This value is considerably smaller than the saturation moment of 3.27 μ_B/Nd theoretically expected for a free Nd ion [15]. At 10, 30 and 50 K the magnetization isotherms show linear tendency. Quite comparable magnetization isotherm was noted for the high-pressure prepared R1212Nb and R1222Nb niobio-cuprates, though they showed different results on magnetic susceptibilities, specific heat measurements as well as on transport properties [10, 11, 16, 17].

We also measured M(*H*) curves for *Tb*$Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound at different temperatures with higher applied fields up to ±70 kOe. The data collected at 1.8 K M(H) curve is shown in the main panel of Fig. (**5**). The low field (±10 kOe) MH plot for the same is also shown in the inset-1 of Fig. (**5**). Like *Nd*/1212Nb compound, the M(*H*) curves measured at 10, 20, 30 and 50 K exhibit linear behavior as confirming the paramagnetic state/component (see Inset-2 of

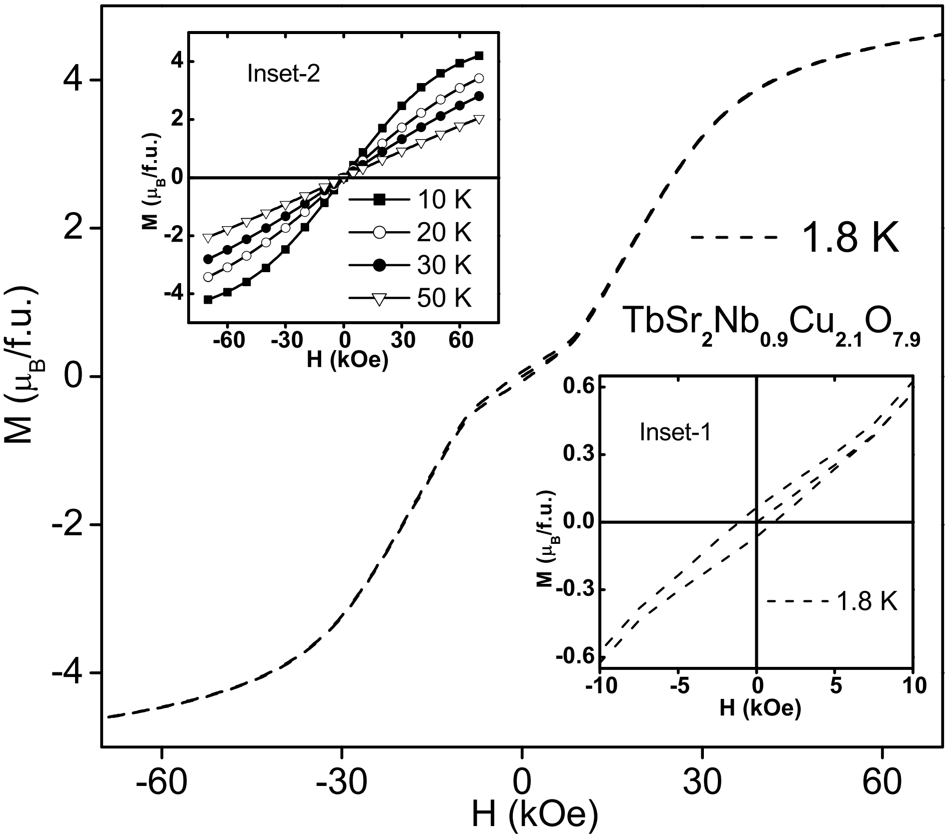

Fig. (5). (main panel) M(H) curve recorded at 1.8 K in higher applied fields up to ± 70 kOe for *Tb*$Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound. Inset-1 in Fig. (**5**) shows the enlarged M(*H*) plot for *Tb*$Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound measured at 1.8 K in fields up to ± 10 kOe with slight loop opening with the coercive field (H_c) of 1.13 kOe and the remnant magnetization (M_r) of about 0.066 μ_B/f.u. Inset-2 in Fig. (**5**) indicates M(H) curves measured at selected temperatures (T = 10, 20, 30, 50 K) in higher applied fields up to ± 70 kOe (–70 kOe ≤ H ≤ 70 kOe) for *Tb*$Sr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound.

Fig. (**5**)). Interestingly, the M(*H*) behavior measured at low temperatures (1.8 K) and ±70 kOe conditions reveals abnormal magnetization/metamagnetism for *Tb*/1212Nb compound. We believe that it is due to spin-flop transition, which is clearly seen in the isostructural Tb-based irido-cuprate compound, $IrSr_2TbCu_2O_8$ [12]. As the field increases the magnetization increases (linearly) with applied field until ~ ±10 kOe with slight loop opening with the coercive field (H_c) of 1.13 kOe and the remnant magnetization (M_r) of about 0.066 μ_B/f.u. (inset-1 of Fig. **5**). It is clear from the inset-1 of Fig.5 that a weak negative susceptibility signal seen at 2 K in the low field (10 Oe) χ(T) measurement is not reflected in the M(H) curve measured at 1.8 K in the low field regime ~ ±10 kOe. Beyond this field regime (~ ±10 kOe) the increase in magnetization is steady and faster than former regime and the effective moment is not saturated even at higher applied field (70 kOe) and low temperature suggesting canted antiferromagnetic interaction in this title compound. The step-like abnormal magnetization isotherm has been reported in *Tb*-containing and some other compounds in literature [18-22]. The magnetization (non-saturated), M ($\mu_B/f.u.$) value estimated at H = 70 kOe for the R = Tb compound (Table **2**) is 4.18 $\mu_B/f.u.$ which is significantly less than the theoretically expected saturation magnetization of the free ion value, 9.0 μ_B/Tb [14]. The reduction in saturation magnetization seen for R = Nd, Tb compounds may be attributed to the crystal field effect. We stress high field magnetization, M(H) isotherm measurements for the presently studied compounds in order to estimate the real saturation moment of the compounds.

4. SUMMARY

In summary, the 1212-type *niobio-cuprates* with starting composition, $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$; R = Nd, and Tb have been prepared through high-pressure solid state reaction route. All the *as*-prepared samples were systematically investigated by XRD, and *dc*-magnetization (χ-T, χ^{-1}(T) and M(*H*)) measurements. The *XRD* patterns of $RSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$; R = Nd, and Tb compounds at room temperature clearly indicate that they are crystallized in tetragonal structure. The χ against T measurements on $NdSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound revealed paramagnetic-nature where as the magnetization versus field, M(*H*) data taken at 1.8 K indicate non-linear increase of magnetization with applied field. Fascinatingly, unlike $NdSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound, the $TbSr_2Nb_{0.9}Cu_{2.1}O_{7.9}$ compound (containing with secondary phase) indicated two antiferromagnetic-like transitions (T_N) at ~41 K as well as at ~ 8 K. The M(*H*) plot measured at 1.8 K and ±70 kOe conditions revealed step-like magnetization isotherm for *Tb*/1212Nb compound. The p_{eff} for the present high-pressure synthesized *Nd*/1212Nb sample (3.71 μ_B/Nd) is slightly larger than the reported ambient-pressure prepared *Nd*/1212Nb (3.1 μ_B/Nd) sample. The negative sign of Weiss paramagnetic temperature, θ_w establishes antiferromagnetic interactions in these studied compounds. Varying the synthesis conditions and controlling the oxygen content under high-pressure may yield phase-pure *Tb*-based 1212-type samples. Further studies on sample optimization and chemical analyses on *Tb*-based sample are warranted.

ACKNOWLEDGEMENTS

The author is thankful to Dr. E. Takayama-Muromachi and NIMS for the use of world class research facilities for the present work.

REFERENCES

[1] Zandbergen HW, Cava RJ, Krajewski JJ, *et al*. High resolution electron microscopy study of $Sr_2NdNbCu_2O_8$. J Solid State Chem 1992; 101: 322-30.

[2] Vybornov M, Perthold W, Michor H, *et al*. Synthesis and characterization of compounds $Sr_2RMCu_2O_{8-d}$ (R = Pr, Nd, Sm,Eu, Gd; M = Nb, Ta). Phys Rev B: Codents Matter 1995; 52: 1389-404.

[3] Kopnin EM, Lobanov MV, Abakumov AM, *et al*. Synthesis and properties of $NbM_2RCu_2O_8$ and $TaM_2RCu_2O_8$ phases (M = Ba, Sr: R = Pr, Nd, Sm, Eu and Gd). J Alloys Compd 1996; 241: 63-8.

[4] Brničević N, Bašic I, Planinić P, *et al*. Phase transformation in the system $Sr_2Ln_{1-x}Ce_xMCu_2O_{8+\delta}$ (Ln = rare earth, M = Nb, Ta) and superconductivity. Appl Superconduct 1993; 1(3-6): 519-26.

[5] Greaves C, Slater PR. Nb and Ta substitutions in $YBa_2Cu_3O_{7-x}$ and related phases: Structural characterisation of $La_{1.1}Ba_{1.9}Cu_{2.1}M_{0.9}O_8$ (M = Nb, Ta). Phyisca C 1989; 161: 245.

[6] Babu TGN, Greaves C. The electrical effects of fermi energy changes in $EuSr_2Cu_2NbO_8$ and related phases. Phyisca C 1991; 585: 185-9.

[7] Greaves C, Slater PR. The synthesis of new phases related to ybco by complete replacement of the chain copper sites. IEEE Trans Magn 1991; 27: 1174.

[8] Hellebrand B, Wang XZ, Steger PL, *et al*. Preparation and characterization of new 1-2-1-2 compounds $MA_2RECu_2O_{8-d}$ and $Nb_{1-x}Ru_xSr_2RECu_2O_{8-d}$ (M= Nb or Ta, A = Ba or Sr, and RE = Pr or Sm). J Solid State Chem 1994; 110: 32-5.

[9] Zandbergen HW, Cava RJ, Krajewski JJ, *et al*. HREM on defects in $Sr_2Nd_{1.5}Ce_{0.5}NbCu_2O_{10-\delta}$. Phys C Superconduct 1992; 196(3-4): 252-8.

[10] Balamurugan S, Awana VPS, Takayama-Muromachi E. High-pressure and high temperature synthesis and magnetic characterization of niobio-cuprate $Nb_{0.9}Sr_2YCu_{2.1}O_{7.9}$. J Appl Phys 2007; 101: 09G110.

[11] Balamurugan S. High-pressure synthesis and physical properties of $NbSr_2(Dy_{1.5}Ce_{0.5})Cu_2O_{10}$. Mod Phys Lett B 2009; 23: 903-10.

[12] Dos santos-García AJ, van Duijn J, Saéz-Puche R, *et al*. $IrSr_2TbCu_2O_8$, a high-pressure metamagnetic cuprate: Structure, microstructure and properties. J Solid State Chem 2008; 181(5): 1167-75.

[13] Spaldin NA, Ed. Magnetic materials: Fundamental and device applications. Cambridge University Press 2003; p. 48.

[14] Goodwin TJ, Radousky HB, Shelton RN. Superconducting, magnetic, electronic transport, and structural properties of $(R_{1.5}Ce_{0.5})$ $Sr_2Cu_2NbO_{10}$, R = Pr, Nd, Sm, Eu. Physica C 1992; 204: 212.

[15] Jensen J, Allan R Eds. Rare earth magnetism-structure and excitations. Mackintosh, Clarendon Press-Oxford 1991; p. 57.

[16] Balamurugan S, Yamaura K, Takayama-Muromachi E. High-pressure synthesis and magnetic properties of the niobio-cuprate $Nb_{0.9}Sr_2HoCu_{2.1}O_{7.9}$. Phys C-Superconduct Appl 2008; 468(15-20): 1206-9.

[17] Blackstead HA, Yelon WB, Kornecki M, *et al*. Antiferromagnetism and superconductivity: Cuprate plane magnetic ordering in $YSr_2Cu_{2.1}Nb_{0.9}O_{8-\delta}$. Phys Rev B (Rapid Communications) 2007; 75: 140514(R) (1-4).

[18] Loewenhaupt M, Doerr M, Rotter M. Magnetic field induced ising axis conversion in $Tb_{0.5}Dy_{0.5}Cu_2$ single crystals. Braz J Phys 2000; 30(4): 754-7.

[19] Chakri NE, Guerrioune M, Fillion G. A study of magnetic properties for sputtered amorphous films Tb-Co based alloys. J Eng Appl Sci 2006; 1(3): 248-51.

[20] Jammalamadaka SN, Mohapatra N, Sitikantha D Das. *et al*. Enhancement of positive magnetoresistance following a magnetic-field-induced ferromagnetic transition in the intermetallic compound Tb_5Si_3. Phys Rev B 2009; 79: 060403R.

[21] Jammalamadaka SN, Mohapatra N, Sitikantha D Das, *et al*. Magnetic anomalies in $Gd_6Co_{1.67}Si_3$ and $Tb_6Co_{1.67}Si_3$. J Phys Condens Matter 2008; 20: 425204.

[22] Richard-Plouet M, Vilminot S, Guillot M, *et al*. Canted antiferromagnetism in an organo-modified layered nickel phyllosilicate. Chem Mater 2002; 14: 3829-36.

Received: August 20, 2009 Revised: October 10, 2009 Accepted: October 26, 2009