Investigation of the magnetic ground state of the ordered double perovskite Sr$_2$YbRuO$_6$: Spin-reorientation and arrest of Yb$^{3+}$ moments

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Abstract:

Comprehensive muon spin rotation/relaxation (μSR) and neutron powder diffraction (NPD) studies supported via bulk measurements have been performed on the ordered double perovskite Sr$_2$YbRuO$_6$ to investigate the nature of the magnetic ground state. Two sharp anomalies/transition at $T_{N1}$ ~ 42 K and $T_{N2}$ ~ 36 K have been observed in the static and dynamic magnetization measurements, coinciding with similar anomalies found in the heat capacity data. In order to confirm the origin of the observed phase transitions and the magnetic ground state, microscopic evidences are presented here. An initial indication of long-range magnetic ordering comes from a sharp drop (nearly a factor 2/3) in the muon initial asymmetry and a peak in the relaxation rate near $T_{N1}$. NPD confirms that the magnetic ground state of Sr$_2$YbRuO$_6$ consists of an antiferromagnetic (AFM) structure with interpenetrating lattices of parallel Yb$^{3+}$ and Ru$^{5+}$ moments lying in the $ab$-plane and adopting a A-type AFM structure. Intriguingly, a small but remarkable change is observed in the long-range ordering parameters at $T_{N2}$ confirming the presence of a weak spin reorientation transition primarily associated with a change in the magnetic moment evolution of the Yb$^{3+}$ spins. The temperature dependent behaviour of the Yb$^{3+}$ and Ru$^{3+}$ moments suggests that the 4$d$-electrons of Ru$^{5+}$ play a dominating role in stabilizing the long range ordered magnetic ground state in the double perovskite Sr$_2$YbRuO$_6$ whereas only the Yb$^{3+}$ moments show an arrest at $T_{N2}$.

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I. Introduction

Mixed ruthenates with perovskite-based crystal structures have been receiving considerable attention in recent decades [1,2,3,4,5,6,7,8,9], because of their interesting magnetic properties including the recent discovery of spin-triplet superconductivity in the layered ruthenate Sr$_2$RuO$_4$ [10]. Despite the rarity of 4$d$-based magnetic materials, SrRuO$_3$ has a robust Curie temperature $T_C \sim 165$ K with saturation magnetization value of 1.4 $\mu_B$/Ru [11], while SrRu$_2$O$_6$ exhibits antiferromagnetic ordering at $T_N = 563$ K [12]. Interestingly Sr$_2$YRuO$_6$, which has essentially the same crystal structure as SrRuO$_3$, but with every second Ru substituted by Y, orders in an antiferromagnetic (AFM) structure, with estimates of ordered Ru moments even higher than those of the parent compound. Although the critical temperature is reduced to 32 K (antiferromagnetic ordering, $T_{N1}$) with a second AFM transition $T_{N2} = 24$ K [1,3].

The first detailed study of the $M_2$LnRuO$_6$ ($M =$Ca, Sr, Ba, and Ln = Y, La, Eu) ruthenium perovskites was carried out by Greatrex et al. [13], who determined the crystal structure, measured the temperature dependence of the electrical resistivity and magnetic susceptibility, and the $^{99}$Ru Mössbauer effect (ME) at 4.2 K. They reported that these materials have a monoclinic $P2_1/n$ space group and are magnetically ordered at 4.2 K, with $T_N$ ranging from 12 K for Ca$_2$LaRuO$_6$ to <80 K for Ba$_2$LaRuO$_6$, with hyperfine magnetic fields $B_H$ at the Ru sites between 56 -60 T due to the electronic magnetic ordering [13]. In subsequent years, the antiferromagnetically ordered Ru-based double perovskites, Sr$_2$LnRuO$_6$ (Ln = rare-earth Ho, Tb, Yb, Dy and Lu or Y etc.) were reported to exhibit two magnetic transitions and strong geometrical frustration above the magnetic ordering for some of these systems confirmed via bulk and microscopic measurements [2,3,5,6,9]. Recent neutron diffraction studies for Ln = Y allowed to understand and differentiate the origin of the two magnetic transitions [3] whereas for Ln = Dy, Ho and Tb, the difference between the two magnetic transitions could not be resolved in the neutron diffraction study with the available instrumental resolution limit [5,9]. In Sr$_2$YRuO$_6$, only half of the Ru-layers order magnetically below $T_{N1}$ while the other half (alternatively) reveals short-range ordering. Furthermore, below $T_{N2}$, the system exhibits a fully ordered type-I AFM ground state [3]. The origin of $T_{N2}$ in some of these double perovskites with type-I AFM structure below $T_{N1}$ is still an open question [5,14,15] and the aim of present work is to resolve this enigma for Sr$_2$YbRuO$_6$. Earlier assumptions that the two magnetic transitions in Sr$_2$YbRuO$_6$ are due to the ordering of Yb and Ru moments at different temperatures seems inadequate and unlikely due to the presence of two such transitions as well in the Ln = Y system where only one magnetic cation (i.e. Ru) is present [3,4]. Further intriguing facts regarding the magnetic ground state of the Ru- based double perovskites are the similar ordered moment values (~2 $\mu_B$) found for the Ruthenium ion irrespective of the nature of the Ln atom and the small value of the ordered moments of the magnetic Ln = rare-earth ions [3,5,9]. The recent $\mu$SR and neutron diffraction results on Ln = Dy suggest the governing role of the Ru-sublattice in inducing the simultaneous ordering of the Dy spins near $T_{N1}$ (40 K) [9]. Also, the detail neutron diffraction study on Ln = Ho and Tb confirms that Ru-O-Ln interactions are mainly responsible for the Ln ordering in the presence of weak super-exchange Ln-O-O-Ln interactions [5]. All these results motivate further to explore the other members of this family in order to understand the origin of the two magnetic transitions, the role of the Ru-atom in the magnetic ordering, and the participation of rare-earth atom in determining the magnetic ground state.

Sr$_2$YbRuO$_6$ is a magnetic insulator with a double-perovskite structure which undergoes a long-range magnetic ordering transition below $T_{N1}$ (42 K), in addition to the conspicuous occurrence of the second transition at $T_{N2} = 36$ K and a weak anomaly at $T^* = 10$ K, consistent with the literature [4,8,14]. Sr$_2$YbRuO$_6$ also displays a temperature induced magnetization
reversal due to an underlying magnetic compensation phenomenon [8]. The observed magnetic entropy \( S_{\text{mag}} = 5.7 \text{ J mol}^{-1} \text{ K}^{-1} \) at 60 K in \( \text{Sr}_2\text{YbRuO}_6 \) is smaller than the expected for the ordered Ru\(^{5+}\) moments with a ground state of \( J = 3/2 \) \( (S_{\text{mag}} = 11.52 \text{ J mol}^{-1} \text{ K}^{-1}) \) [4] which could be due to the presence of the large crystal field (CEF) of Ru and due to the CEF effect of rare-earth atom (Yb). The same group has also reported the exchange bias effect in \( \text{Sr}_2\text{YbRuO}_6 \) below the compensation temperature [16]. The compensation temperature was referred as the temperature where the measured magnetization becomes zero [4] and a cross-over of zero field cooled and field cooled magnetization occurs. However, in the same report, it was suggested that two magnetic anomalies near \( T_{N1} \) and \( T_{N2} \) could be due to the magnetic ordering of Ru\(^{5+}\) and Yb\(^{3+}\) moments, respectively. Later, Doi et al. [15] reported a type-I AFM structure below \( T_{N1} \) confirmed via NPD study performed at 10 K. However, due to a lack of a systematic temperature dependent NPD data, no information is available regarding the thermal evolution of magnetic structure at \( T_{N2} \) [14]. Here, we present a detailed study which confirms that both the Ru\(^{5+}\) and Yb\(^{3+}\) moments order at \( T_{N1} \) and that a weak spin reorientation takes place at \( T_{N2} \). No change or anomaly has been found near \( T^* \approx 10 \text{ K} \) in NPD data. It is worthy to mention that the presence of 10 K anomaly is still suspicious and one can always argue that it could be due to minute magnetic impurity present in the sample. However, with the available NPD data used in the present study, we are unable to trace any impurity except Yb\(_2\)O\(_3\) which couldn’t explain the 10 K anomaly as discussed in the result section.

II. Experimental details

The polycrystalline sample of \( \text{Sr}_2\text{YbRuO}_6 \) was prepared by the standard solid-state reaction using the same protocol as mentioned elsewhere [4,8]. Phase purity was confirmed by X-ray diffraction (XRD) using Rigaku Smartlab X-ray diffractometer (XRD) equipped with Ge two bounce monochromator enabling Cu-K\(\alpha\) radiation. The \( dc \) magnetization measurements have been performed on Quantum Design’s SQUID magnetometer. Temperature dependent heat capacity, using a relaxation technique, and \( ac \)-susceptibility measurements were performed by Quantum Design, PPMS. To investigate the magnetic structure/ground state, temperature dependent NPD measurements were carried out using the time-of-flight diffractometer WISH at the ISIS Facility, UK [17]. The Fullprof_Suite has been used to analyse the XRD and NPD data [18]. The MuSR spectrometer in longitudinal geometry at the ISIS Pulsed Neutron and Muon Source, UK has been employed to carry out zero-field (ZF) \( \mu \)SR experiments. The powder sample was mounted onto a silver plate (99.999% purity) using GE-varnish and was covered with thin silver foil. The \( \mu \)SR measurements were carried out using He\(^4\) cryostat between 2 and 300 K.

III. Results and discussion

Room temperature XRD pattern of \( \text{Sr}_2\text{YbRuO}_6 \) have been Rietveld refined using monoclinic symmetry (space group \( P2_1/n \)) with an ordered arrangement of Yb\(^{3+}\) and Ru\(^{5+}\) atoms at B-site. The result is shown in Fig. 1 and it is in good agreement with the existing literature [4,8,14]. No extra peaks were evident in the XRD pattern while a very minute impurity phase of Yb\(_2\)O\(_3\) was evident in NPD pattern. One can easily miss out the minute impurity with lab source based XRD machine while the high intensity available on WISH instrument, such minute phase can be easily seen. The results of NPD are discussed in the later sections.

Figure 2(a) displays the zero-field cooled (ZFC) and field cooled (FC) \( dc \) magnetization \( (\chi_{dc}) \) behaviour of \( \text{Sr}_2\text{YbRuO}_6 \) measured in different fields namely, at 50 Oe, 100 Oe and 10 kOe as a function of temperature. The bifurcation starts only below a certain critical temperature, following by a crossover between the ZFC and FC curve. For lower applied fields (50 and 100 Oe), the FC magnetization becomes negative by cooling the sample below the
crossover point, whereas for sufficiently higher field like 10 kOe, only a positive value of FC curve has been observed. Also, for sufficiently higher field, the FC value is higher than the ZFC curve, unlike the low field data. Noticeably, the ZFC magnetization decreases below 42 K showing a plateau for a small temperature region down to 36 K. Below 36 K, it increases with decreasing temperature, irrespective of applied field value. Here we denote these anomalies by $T_{N1}$ (42 K) and $T_{N2}$ (36 K), respectively. The justification and microscopic evidence to denote them as AFM ordering (Néel) temperatures comes from the NPD results which are discussed later. Another intriguing feature is the presence of a weak anomaly near 10 K. A similar anomaly below 15 K was also pointed out previously by Singh et al. [4]. For instance, we denote this by $T^*$, as we don’t have any existing information about its origin. Both the ZFC and FC curves exhibit a small kink near $T^*$.

To investigate in more detail the magnetic anomalies, isothermal magnetization has been measured at selected temperatures across these anomalies starting from 300 K. Figure 2(b) represents the magnetic isotherms measured at T = 5, 30, 37, 50 and 300 K. Noticeably, a weak hysteresis starts to develop below 37 K and becomes quite prominent for the 5 and 30 K curves. It suggests the contribution of a minor ferromagnetic component to the dominant AFM ground state. The 300 K curve exhibits a linear behaviour, as expected from the paramagnetic state. The curve for 50 K also shows a linear behaviour but the magnetization value is larger at 300 K. Furthermore, our detailed dc magnetization study evidences three magnetic anomalies/transition namely at $T_{N1} = 42$ K, $T_{N2} = 36$ K and very weak at $T^* \sim 10$ K.

To understand the dynamic response of these anomalies, ac susceptibility ($\chi_{ac}$) of Sr$_2$YbRuO$_6$ has been measured. Figure 3 represents the real ($\chi'$) part of $\chi_{ac}$ as a function of temperature measured at different frequencies. Two clear anomalies are visible in $\chi'$ behavior near $T_{N1}$ and $T_{N2}$. The frequency independent behaviour of the first anomaly at $T_{N1}$ indicates the onset of long-range ordering below $T_{N1}$. A weak frequency dispersion can be seen near $T_{N2}$ which indicates the change in magnetic interactions at this point. However, in the light of other microscopic evidences given in the next section, we cannot interpret this dispersion as the signature of short-range ordering. A similar kind of frequency dispersion has also been observed even for systems having long range ordered state, i.e. for Sr$_3$NiIrO$_6$ and Sr$_2$DyRuO$_6$ (near $T_{N2}$). [9,19,20]. A very weak, indirect but apparent signature of a third anomaly near $T^*$ can be seen in the $\chi_{ac}$ behavior. The frequency dispersion decreases below $T^*$ and $\chi'$ increases sharply, concomitant with $\chi_{dc}$. The direct signatures of $T_{N1}$ and $T_{N2}$ have been also found in $\chi''$ behavior but due to the weak signal, it is difficult to find any signature of $T^*$ in $\chi''$ behavior (data is not shown here).

The heat capacity of Sr$_2$YbRuO$_6$ measured in 0 and 2 Tesla applied field is presented in Fig. 4. Two clear peaks are visible near 42 K and 36 K, coinciding with the magnetic anomalies at $T_{N1}$ and $T_{N2}$, respectively which confirms the long-range ordering at these transitions. However, no feature or anomaly has been observed near $T^*$. Also, there is no appreciable change in the heat capacity behaviour measured with 0 and 2 T applied field (Fig. 4). Therefore, the static and dynamic magnetization and heat capacity measurements confirm the presence of two long-range transitions at $T_{N1}$ and $T_{N2}$.

In order to understand the microscopic origin and local magnetic response of different phase transitions as observed through the bulk techniques, the zero field (ZF) $\mu$SR spectra of Sr$_2$YbRuO$_6$ have been recorded at various temperatures between 2 and 90 K as shown in Fig. 5. The spectra at 90 and 50 K exhibit weak relaxation and have the same initial asymmetry. However, below 45 K, the relaxation rate increases faster and the initial asymmetry decreases with decreasing temperature, which is a typical behaviour observed near a long-range
magnetic ordering transition. The ZF $\mu$SR data is fitted using an exponential function with a constant background.

$$G_{z}(t) = A_0 \exp(-\lambda t) + A_{bg}$$

(1)

Here $A_0$ is the muon initial asymmetry, $\lambda$ the muon relaxation rate, $A_{bg}$ is the constant background arising from muons stopping on the sample holder. The value of $A_{bg}$ was estimated from fitting the 90 K data and then it was kept fixed for fitting the data at other temperatures. The fitting parameters, relaxation rate ($\lambda$) and initial asymmetry ($A_0$) are plotted in Fig. 6. For temperatures down to 50 K, the initial asymmetry is almost temperature independent, which can be attributed to paramagnetic moment fluctuations of the Yb$^{3+}$ and Ru$^{5+}$ ions. The $\lambda(T)$ increases below 50 K and exhibits a sharp maximum near 42 K ($T_{N1}$). At $T_{N1}$, the initial asymmetry drops down to more than 2/3 of initial value which indicates that the magnetic ordering is bulk in nature. In a polycrystalline sample, below the magnetic ordering temperature muons see three components (one longitudinal and two transverse) of the internal field at muon stopping sites. For a bulk magnetic ordering with larger magnetic moments one expects 2/3 loss of initial asymmetry (2/3 transverse component gives oscillations and 1/3 longitudinal component gives relaxation) as the transverse component can be seen only very close to zero-time limit for larger internal fields at muon stopping sites. In the present case, the asymmetry loss is slightly more than 2/3, which could be due to a fast component below $T_{N1}$ at smaller time, which cannot be estimated due to the muon pulse width at ISIS. For $T < T_{N1}$, the further loss in initial asymmetry is very small and the $\lambda(T)$, after peaking at $T_{N1}$, continues to decrease down to 10 K and below it $\lambda(T)$ decreases faster. $A_0$ does not reveal any sign of a second/third transitions, which one would expect, as the system is in a complete long-range magnetic order state below $T_{N1}$ and hence cannot lose further asymmetry. It is interesting to compare that the observed maxima/peak in $\lambda(T)$ near $T_{N1}$ agrees with the susceptibility and heat capacity data.

This result indicates that within the $\mu$SR time window the first transition at $T_{N1}$ is due to static long-range magnetic ordering of both Yb$^{3+}$ and Ru$^{5+}$ moments, as also observed in the heat capacity and magnetization measurements. However, the continuous change in $\lambda(T)$ across $T_{N2}$ and $T^*$ indicates small changes in the spin structure specifically at $T_{N2}$. Similar kind of responses have been recently observed for various other perovskites [7,21,22,23,24] and have been helpful in exploring the magnetic ground states, including the microscopic co-existence of magnetic ordered and non-magnetic phases in Ba$_2$PrRu$_{0.9}$Ir$_{0.1}$O$_6$ using $\mu$SR [25].

To investigate the magnetic ground state and the possible changes in the magnetic structure across the different transitions, NPD data have been collected on the WISH time-offlight diffractometer at several temperatures between 100 to 2 K with close data points between 45 and 2 K. Emergence of new peaks has been clearly observed below $T_{N1}$. Fig. 7 represents the 3D plot of magnetic Bragg reflections observed for $T < 42$ K and interplanar spacing $d > 3.5$ Å. All the magnetic reflections could be indexed with propagation vector $k = (0,0,0)$. The occurrence of the (010) reflection indicates that the magnetic moments may have components either in the $a$ or $c$-direction. No new peak emerges further below $T_{N2}$ and $T^*$. Each observed reflection exhibits a small but visible enhancement in the intensity which suggests a change in magnetic structure across $T_{N2}$. The black arrows in Fig. 7 point to the temperature region of the diffraction patterns corresponding to $T_{N2}$. Fig. 8 represents the thermal evolution of the intensity of various magnetic reflections observed after subtracting the nuclear contribution. All the peaks exhibit a sharp rise below $T_{N1}$ concomitant with the onset of long-range ordering. Below $T_{N2}$, they exhibit an accelerated enhancement in the diffracted intensity with decreasing temperature, as can be evidenced as well by eye in Fig. 7. Since, all the observed magnetic Bragg peaks can be fitted with the type-I AFM structure (which is discussed below in detail), the observation of two different temperature regions in the thermal behavior of the magnetic
reflections can explain the existence of two peaks in the magnetization and the heat capacity behavior. The red lines in Fig. 8 are guides to the eye for the expected temperature variation. The temperature evolution of (010) and (100/001) peaks in Fig. 8 (a and b) clearly supports the presence of two magnetic transitions. On further cooling below \( T_{N2} \), the intensity keeps on increasing before nearly saturating to some value. Therefore, no further deviation or anomalous change in the long-range order parameter has been observed at \( T^* \). The similar kind of behavior was also observed for \( \text{Sr}_2 \text{YRuO}_6 \) [3].

To investigate the corresponding changes in the magnetic structure of \( \text{Sr}_2 \text{YbRuO}_6 \), Rietveld refinements have been performed of the pristine as well as of the temperature dependent difference data sets where the nuclear contribution using the 45 K data set had been subtracted. Fig. 9 represents the Rietveld refined plot of 100 K (a) and 2 K (b) data from the bank 2 of WISH instrument. All the five banks data have been refined simultaneously to get the final parameters. To probe the smaller changes in magnetic structure at \( T_{N2} \), the temperature difference data have been also refined which are more sensitive for the small changes expected in the magnetic structure at \( T_{N2} \). The 100 K data are fitted with single nuclear phase having monoclinic space group \( P\overline{2}1/n \). A very minute (1.5 \%) impurity of \( \text{Yb}_2\text{O}_3 \) which orders at 2.25 K [26], was found in NPD pattern. The 2 K data are fitted with two-phase (nuclear + magnetic) model. The inset in Fig. 9b shows the Rietveld refined plot of temperature difference data at 2 K (2 K-45 K) which are fitted with single magnetic phase. The collinear model, having parallel \( \text{Yb}^{3+} \) and \( \text{Ru}^{5+} \) moments, has been used to refine the data for the magnetic structure determination. The refined lattice parameters at 2 K are \( a = 5.7305(2) \) Å, \( b = 8.1021(3) \) Å, \( c = 5.7360(2) \) Å and \( \gamma = 90.20(2) \)°. It should be noticed here that we have used the \( P112_1/n \) setting instead of the standard \( P12_1/n1 \), because the former gives an advantage to adopt the polar coordinates during the refinement procedure. The empirically determined magnetic form factor of Ru\(^{5+}\) has been used for the refinement [27]. The magnetic symmetry analysis was performed using the space group \( P\overline{2}1/n \) with \( k = (0, 0, 0) \) using the program BASIREPS [28] which generates two possible irreducible representations (IR1 and IR2), each containing three basis vectors. IR1 has ferro- (F) coupling along the \( c \)-direction and antiferro- (AF) coupling in the \( a \) and \( b \)-directions while on the contrary, the IR2 has AF coupling in the \( c \)-direction and F-coupling in the \( a \) and \( b \)-directions. The best fit of the data can be achieved with a single IR1, having AF-coupling along the \( a \) and \( b \)-direction. The final magnetic structure is presented in Fig. 10 which consists of an interpenetrating lattice of canted moments of \( \text{Yb}^{3+} \) and \( \text{Ru}^{5+} \) ions where ferromagnetic sheets are coupled antiferromagnetically along the \( b \)-direction and ferromagnetically along the \( a \)-direction. An equally good fit of the data can be obtained by refining the magnetic structure with \( b \) and \( c \) components, due to pseudosymmetry present in the sample. However, owing to the monoclinic symmetry it is more likely that the moments will align in the \( ab \)-plane rather than the \( bc \)-plane. Doi et al. [15] have reported the moments to be in the \( bc \) plane with a 23° canting angle at 10 K, according to their choice of lattice parameters, where \( a < b < c \) and \( \gamma \) represents the monoclinic distortion. It is important to note here that due to pseudosymmetry between the \( a \) and \( c \)-parameters, it is not possible to distinguish the \( a \) and \( c \)-component. The direction of the magnetic moments of \( \text{Yb}^{3+} \) and \( \text{Ru}^{5+} \) are different in the present system from those of the Ho, Tb and Dy based double ruthenates [5,9]. The coupling between the \( \text{Yb}^{3+} \) and the \( \text{Ru}^{5+} \) moments is ferromagnetic whereas an antiferromagnetic coupling was observed between \( \text{Dy}^{3+} \) and \( \text{Ru}^{5+} \) moments in \( \text{Sr}_2\text{DyRuO}_6 \) [9]. Also, the spins are aligned along the long \( b \)-axis with an angle of \( \sim 45-51^\circ \) (temperature dependent) with respect to the \( a \)-axis. The values of the \( \text{Yb}^{3+} \) and the \( \text{Ru}^{5+} \) moments at 2 K are \( \mu_{\text{Yb}^{3+}} = 0.536(7) \mu_B \) and \( \mu_{\text{Ru}^{5+}} = 2.095(7) \mu_B \). The strong reduction of \( \mu_{\text{Yb}^{3+}} \) compared to the expected value of \( \sim 4.5 \mu_B \) matches with similar discrepancies observed for \( R = \text{rare-earths} \) moment for the other members of Ru-based perovskites family, like \( \text{Sr}_2\text{DyRuO}_6 \), \( \text{Sr}_2\text{HoRuO}_6 \), \( \text{Sr}_2\text{YRuO}_6 \), etc.
and Sr₂TbRuO₆ etc. [5,9,15,29], which could be due to the effect of the crystal field on the rare-earth cation. Symmetry allows in theory a ferromagnetic component in IR1 on the Yb³⁺ and the Ru⁵⁺ moments along the c-direction. In the NPD data the possible ferromagnetic component would come on top of the strong nuclear peaks making the detection of any weak ferromagnetic component very difficult. Table I contains the information on the bond lengths and bond angles variation during these transitions. No sudden change or variation in the bond lengths/bond angles was noticed during $T_{N2}$ or $T^*$. To discern the change in magnetic structure at $T_{N2}$, the temperature variation of the magnetic moments has been estimated by performing the Rietveld refinement of the temperature dependent difference data sets. The refined Ru⁵⁺ and Yb³⁺ moments are plotted in Fig. 11 as a function of temperature alongside with the angle $R_0$ with respect to the x-axis (a-axis) and the normalized moments. There are clear but small anomalies in the temperature dependence of the moments (more pronounced for Yb³⁺) and in the $R_0$ value at $T_{N2}$. The Yb³⁺ moments show a sharp increase (similar to the Ru⁵⁺ moments) below $T_{N1}$, but show an arrest in the slope near $T_{N2}$, below it increasing again more strongly (almost linearly with temperature) and saturating near 10 K. The angle $R_0$ which is determined by the relative amounts of the two antiferromagnetic components along the $a$- and the $b$-directions also shows a continuous increase down to $T_{N2}$, below which it slightly decreases before saturating to ~50°. Also, from Fig. 11d, it appears that below $T_{N2}$ the Ru⁵⁺ moments attain the saturation value with much faster rate as compared to Yb³⁺. Noticeably, the rate of increase of the Ru⁵⁺ and Yb³⁺ moments is different only below $T_{N2}$ while both the moments increase with same rate between $T_{N1}$ and $T_{N2}$. Resuming the analysis of the temperature dependent refinement of the difference data sets one can say that a broad but clear peak in $R_0$ along with a small plateau in Yb³⁺ moments near $T_{N2}$ and the temperature variation of normalized moments confirm the spin reorientation transition at $T_{N2}$ in Sr₂YbRuO₆, supported by the change in magnetic peak intensity as discussed above.

It should be noted that the Ru⁵⁺ moment value for the present case is very similar to the reported values for other members of this double perovskite family and points to the fact that in these systems the Ru-O-O-Ru interactions are the strongest magnetic interactions, which control the Ru ordering [3,5,9,15,29]. The very low value of $T_N = 2.3$ K for rare-earth oxide Yb₂O₃ indicates that Yb-O-Yb interactions are in general very weak [26]. For the well-ordered double perovskite Sr₂YbRuO₆, only weaker super-super exchange Yb-O-O-Yb interactions will be present. These interactions cannot be solely responsible for the Yb³⁺ ordering at $T_{N1}$. Therefore, it appears that the Ru-O-Yb interactions have an important role in governing the magnetic ordering of the rare-earth cation Yb³⁺. Noticeably, the Yb³⁺ moment exhibits deviation from mean field type behavior as a function of temperature while the Ru⁵⁺ moment follows the mean field behaviour down to 2 K. This indicates that in the rare-earth and ruthenium-based perovskites, the primary magnetic ordering below $T_{N1}$ is induced by the order of the 4$d$-electrons of Ru⁵⁺ rather than that the rare-earth cation, as also verified for Sr₂LnRuO₆ (Ln = Ho, Tb and Dy) [5,9], while a spin reorientation of Yb³⁺ spins takes place at $T_{N2}$.

IV Conclusions

We have investigated the ordered double perovskite Sr₂YbRuO₆ using various experimental techniques to understand the origin of reported two magnetic transitions. The bulk magnetization measurements of Sr₂YbRuO₆ reveal the presence of two clear magnetic transitions as a function of temperature, namely at $T_{N1} = 42$ K, $T_{N2} = 36$ K and a very weak anomaly at $T^* = 10$ K. The heat capacity measurements reveal a clear signature of $T_{N1}$ and $T_{N2}$ indicating the long-range ordering whereas no anomaly can be detected at $T^*$. Our detailed µSR and NPD results provided a concrete evidence of long-range magnetic ordering below $T_{N1}$
and a clear change in the long-range magnetic ordering parameters at $T_{N2}$. Interestingly, the magnetic ordering is primarily controlled by the Ru$^{5+}$ moments, but a clear spin-reorientation transition at $T_{N2}$ is confirmed based on the temperature variation of Yb$^{3+}$ and Ru$^{5+}$ moments and the moments angle $R_0$. All the observed magnetic Bragg peaks can be indexed with single propagation vector $\mathbf{k} = (0, 0, 0)$ and the magnetic structure consists of interpenetrating sublattices of Yb$^{3+}$ and Ru$^{5+}$ spins having confined moments in the $ab$-plane. The resultant magnetic structure is composed of parallel spins of Yb$^{3+}$ and Ru$^{5+}$ and having an angle of $\sim 45$-51° with respect to $a$-direction. To understand the origin of third anomaly at $T^*$, further investigations, especially on single crystal of Sr$_2$YbRuO$_6$, are needed. The present results will help to understand the observed two magnetic phase transitions in other double perovskites and will be important to develop a realistic theoretical model to explain the mechanism of two magnetic transitions in double perovskites family.

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Table I: Selected bond angles (°) and bond lengths (Å) in paramagnetic (45 K) and antiferromagnetic state (2 K).

| Bond angles (°) | 45 K       | 2 K       | Bond lengths (Å) | 45 K       | 2 K       |
|----------------|------------|-----------|------------------|------------|-----------|
| O1-Ru-O2       | 90.2(4)    | 90.1(4)   | Ru-O1            | 1.958(8)   | 1.960(9)  |
| O1-Ru-O3       | 90.9(4)    | 90.8(4)   | Ru-O2            | 1.959(9)   | 1.957(1)  |
| O2-Ru-O3       | 89.5(3)    | 89.2(4)   | Ru-O3            | 1.941(8)   | 1.944(8)  |
| O1-Yb-O2       | 87.9(4)    | 91.9(4)   | Yb-O1            | 2.164(8)   | 2.163(9)  |
| O1-Yb-O3       | 90.1(3)    | 89.8(3)   | Yb-O2            | 2.172(9)   | 2.172(1)  |
| O2-Yb-O3       | 89.1(3)    | 89.3(3)   | Yb-O3            | 2.182(8)   | 2.180(8)  |
| Ru-O1-Yb       | 159.1(5)   | 159.0(5)  |
| Ru-O2-Yb       | 157.7(5)   | 158.1(5)  |
| Ru-O3-Yb       | 158.4(5)   | 158.1(5)  |
Figure Captions:

Fig. 1: Rietveld refined XRD pattern of Sr₂YbRuO₆ using monoclinic space group P112₁/n.

Fig. 2: (a) The dc magnetic susceptibility (χ<sub>dc</sub>) measured at various applied magnetic fields in zero-field cooled (ZFC) and field cooled (FC) conditions. The arrows indicate the magnetic transitions as T<sub>N1</sub> and T<sub>N2</sub> and the dashed line indicates the third weak anomaly T* near 10 K. The inset shows the enlarge view close to magnetic transitions. (b) Magnetization isotherms measured at various temperatures ranging from 5 to 300 K. The inset shows the enlarge view at lower fields data to show the hysteresis observed at 5 K and 30 K.

Fig. 3: Real part of ac susceptibility (χ<sub>′</sub>) measured with 10 Oe drive field in zero field cooled conditions at different frequencies ranging from 100 Hz to 10 kHz. The arrows indicate the magnetic transitions at T<sub>N1</sub> and T<sub>N2</sub>. The inset represents the zoomed view around T<sub>N1</sub> and T<sub>N2</sub>.

Fig. 4: Heat capacity as a function of temperature in zero and 2 Tesla applied magnetic field.

Fig. 5: Zero-field μSR spectra measured at various temperatures. The experimental data are shown by the symbols and the solid red line shows fit to the data using an exponential decay function.

Fig. 6: The temperature dependent parameters obtained from the fit to μSR spectra as a function of temperature. The initial muon asymmetry (A₀) and relaxation rate (λ) are plotted on right and left y-scale with linked x-scale.

Fig. 7: Thermal evolution of magnetic reflections below T<sub>N1</sub>. The arrows indicate the magnetic Bragg reflections and highlights the small enhancement in diffracted intensity below T<sub>N2</sub>.

Fig. 8: (a-d) The temperature variation of various magnetic reflections extracted from the difference curve. Two components are clearly visible as shown by the two red lines in Fig. (a, b) whereas a single line is shown in Fig. (c, d) which deviates from the observed intensity pattern for T<sub>N2</sub> > T > T<sub>N1</sub>, as highlighted in the shaded region within the circle. (Refer to the text for details).

Fig. 9: Rietveld refined NPD patterns collected at (a) 100 and (b) 2 K. Two series of tick marks in (b) correspond to the nuclear (upper-green) and magnetic (lower-red) Bragg reflections. The observed, calculated intensities and difference are plotted as solid circles, solid line and bottom line, respectively. The inset in (b) shows the fitted difference data (2 K-45 K) using the same model.

Fig. 10: The magnetic structure of Sr₂YbRuO₆ for k = (0 0 0). The Yb<sup>3+</sup> and Ru<sup>5+</sup> moments are shown in cyan (small) and red colours, respectively.

Fig. 11: Thermal variation of (a) Ru<sup>5+</sup> moments, (b) Yb<sup>3+</sup> moments, (c) moments angle θ<sub>0</sub> (with respect to x-axis/a-axis) and (d) Normalized moments of Yb<sup>3+</sup> and Ru<sup>5+</sup>. The vertical black dashed line corresponds to T<sub>N2</sub>. 
Fig. 1: Rietveld refined XRD pattern of Sr$_2$YbRuO$_6$ at 300 K using monoclinic space group $P112_1/n$.  

$\chi^2 = 1.24$
Fig. 2: (a) The dc magnetic susceptibility ($\chi_{dc}$) measured at various applied magnetic field in zero-field cooled (ZFC) and field cooled (FC) conditions. The arrows indicate the magnetic transitions as $T_{N1}$ and $T_{N2}$ and the dashed line indicates the third weak anomaly $T^*$ near 10 K. The inset shows the enlarge view close to magnetic transitions. (b) Magnetization isotherms measured at various temperatures ranging from 5 to 300 K. The inset shows the enlarge view at lower fields to show the hysteresis observed at 5 K and 30 K.
Fig. 3 Real part of \( ac \) susceptibility (\( \chi' \)) measured with 10 Oe drive field in zero field cooled conditions at different frequencies ranging from 100 Hz to 10 kHz. The arrows indicate the magnetic transitions at \( T_{N1} \) and \( T_{N2} \). The inset represents the zoomed view around \( T_{N1} \) and \( T_{N2} \).

Fig. 4: Heat capacity as a function of temperature in the presence of 0 and 2 Tesla applied magnetic field.
Fig. 5: Zero-field μSR spectra measured at various temperatures. The experimental data are shown by the symbols and the solid red lines show the fit to the data using an exponential decay function.
Fig. 6: The temperature dependent parameters obtained from the fit to μSR spectra as a function of temperature. The initial muon asymmetry ($A_0$) and relaxation rate ($\lambda$) are plotted on right and left y-scale with linked x-scale.

Fig. 7: Thermal evolution of magnetic reflections below $T_{N1}$. The arrows indicate the magnetic Bragg reflections and highlights the small enhancement in diffracted intensity below $T_{N2}$. 
Fig. 8: (a-d) The temperature variation of various magnetic reflections extracted from the difference curve. Two components are clearly visible as shown by the two red lines in Fig. (a, b) whereas a single line is shown in Fig. (c, d) which deviates from the observed intensity pattern for $T_{\text{N2}} > T > T_{\text{N1}}$, as highlighted in the shaded region within the circle. (Refer to the text for details).
Fig. 9: Rietveld refined NPD patterns collected at (a) 100 and (b) 2 K. Two series of tick marks in (b) correspond to the nuclear (upper-green) and magnetic (lower-red) Bragg reflections. The observed, calculated intensities and difference are plotted as solid circles, solid line and bottom line, respectively. The inset in (b) shows the fitted difference data (2 K- 45 K) using the same model.
Fig. 10: The magnetic structure of Sr$_2$YbRuO$_6$ for $k = (0\ 0\ 0)$. The Yb$^{3+}$ and Ru$^{5+}$ moments are shown in cyan (small) and red colours, respectively.
Fig. 11: Thermal variation of (a) Ru$^{5+}$ moments, (b) Yb$^{3+}$ moments, (c) moments angle $R\theta$ (with respect to x-axis/a-axis) and (d) Normalized moments of Yb$^{3+}$ and Ru$^{5+}$. The vertical black dashed line corresponds to $T_{N2}$. 
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