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Synthesis and Characterization of the Double Perovskite Y_2MgRuO_6

Sahana Rößler , Ulrich Burkhardt, Liu H. Tjeng and Alexander C. Komarek * 

Max Planck Institute for Chemical Physics of Solids, Nöthnitzer Str. 40, D-01187 Dresden, Germany; sahana.roessler@cpfs.mpg.de (S.R.); ulrich.burkhardt@cpfs.mpg.de (U.B.); tjeng@cpfs.mpg.de (L.H.T.)

* Correspondence: alexander.komarek@cpfs.mpg.de; Tel.: +49-351-4646-4423

Abstract: A new double perovskite compound with nominal composition Y_2MgRuO_6 is synthesized by the floating zone technique. X-ray diffraction measurements reveal a monoclinic crystal structure with space group $P2_1/n$ and monoclinic angle $\beta = 90.178(1)^\circ$ as crystallographically expected for a double perovskite. A composition of $\text{Y}_{1.880(8)}\text{Mg}_{0.953(4)}\text{Ru}_{1.047(4)}\text{O}_{6-\delta}$ is revealed by our single crystal X-ray diffraction measurement. The magnetic susceptibility of this compound does not show indications of magnetic ordering down to the lowest temperature. A Curie–Weiss fit for the paramagnetic part above 300 K yields an effective moment that indicates an $S = 1$ Ru^{4+} state. We attribute the occurrence of the $S = 1$ state and the lack of magnetic order to the presence of the B/B' -site disorder.

Keywords: floating zone method; double perovskite; ruthenate; crystal structure; magnetization

1. Introduction

Ru^{4+} materials with a $4d^4$ configuration have recently attracted considerable interest due to theoretical models that propose unconventional magnetism arising from an interplay of spin–orbit coupling (SOC), superexchange and electron correlations [1–3]. Large crystal field and SOC in these materials might lead to a $J = 0$ ground state, which is expected to show only temperature-independent Van Vleck-type magnetism. In cubic K_2RuCl_6 , the $J = 0$ ground state has been observed, indicating the unquenched orbital angular momentum in this compound [4]. In this type of material, the superexchange energy scale is comparable to the SOC, and therefore excitations between the $J = 0$ ground state and $J = 1$ triplon excited states are possible [5]. And new interesting magnetic ground states might appear in these materials [1–3,5–7]. Several Ru^{4+} systems have been already studied in the past [8–13].

Compounds with a double perovskite structure are interesting because a face-centered cubic structure could induce bond-dependent frustration in spin–orbit coupled excitonic states [5,14]. In order to investigate the magnetic properties related to isolated Ru^{4+} ions in the perovskite structure, a B -site ordered structure with Ru as the only magnetic ion is necessary. Several Ru^{4+} double perovskite compounds have been already synthesized [15]. For La_2MRuO_6 (with the M^{2+} -ions Mg^{2+} , Co^{2+} , Ni^{2+} and Zn^{2+}), a double perovskite structure has been found [15–17]. (B -site ordering depends on the synthesis conditions [15,18,19]). For $\text{La}_2\text{MgRuO}_6$, the effective paramagnetic moment μ_p was found to be somewhat larger than calculated for a spin-only $S = 1$ localized $t_{2g}^4 e_g^0$ electron configuration for a low spin state: $\mu_p = 3.22 \mu_B$ and $\Theta = -100$ K [15,20]. Possibly, the formation of Ru-4d bands could be responsible for these deviations [15,20]. Also for $\text{La}_2\text{ZnRuO}_6$, a similar behavior with $\mu_p = 3.24 \mu_B$ and $\Theta = -80$ K was observed [20]. For both compounds, the magnetic susceptibility deviates from Curie–Weiss behavior below about 250 K [20].

Here, we report the crystal growth, structure, and properties of a new double perovskite with the nominal composition Y_2MgRuO_6 that cannot be synthesized at an ambient pressure and high-temperature conditions.



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2. Results and Discussion

Single crystals of Y_2MgRuO_6 were grown by the floating zone technique—see Figure 1a for a Laue diffraction pattern of an as-grown crystal. The starting materials Y_2O_3 , RuO_2 and MgO were mixed in stoichiometric amounts with 100% RuO_2 excess for a compensation of RuO_2 evaporation (see also other ruthenate growth reports, e.g., ref. [21] and references therein). This mixture was thoroughly ground and pressed into rods of ~ 5 mm in diameter. These rods were sintered in air at 1100°C for 24 h. The subsequent floating zone growth was performed at a growth rate of 15 mm/h under an O_2/Ar atmosphere of 10 bar (with a ratio of 1:10) at a temperature of about 1800°C . Despite the large excess of RuO_2 , the fast growth rate was still necessary to cope with the heavy evaporation of RuO_2 during crystal growth.

Attempts to synthesize polycrystalline Y_2MgRuO_6 samples using a conventional solid state synthesis route failed. For these polycrystalline samples, stoichiometric amounts of Y_2O_3 , MgO , and RuO_2 were mixed and first reacted at 950°C for 24 h followed by sintering at 1200°C for 48 h. However, powder X-ray diffraction measurements did not show any evidence for a Y_2MgRuO_6 phase in these powder samples. Instead, an $\text{Y}_2\text{Ru}_2\text{O}_7$ pyrochlore phase [22] was found, together with impurity phases of Y_2O_3 and MgO .

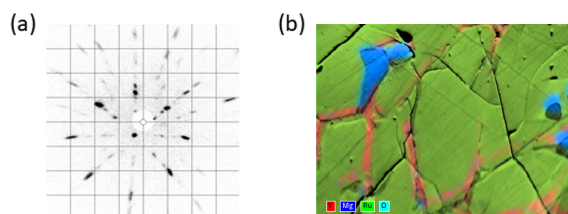


Figure 1. (a) Laue diffraction pattern of an as-grown Y_2MgRuO_6 crystal. (b) EDX map showing inclusions of Y_2O_3 and MgO assigned by EBSD pattern evaluation.

An electron back-scatter diffraction (EBSD) measurement of a part of the as-grown boule shows a homogeneous phase distribution with grains of the monoclinic Y_2MgRuO_6 majority phase and the Y_2O_3 and MgO phases at the grain boundaries, see Figure 1b. It was possible to extract single crystalline grains of up to ~ 30 – $40\ \mu\text{m}$ dimension from the floating zone grown crystal, which could be used for further measurements.

The composition of the as-grown polycrystalline material was analyzed using wavelength dispersive X-ray (WDX) on an electron microprobe. The following ratios were obtained for the atomic concentrations of the metals— $\text{Y}:\text{Mg}:\text{Ru} = 2.00(1):0.85(1):1.11(1)$. The oxygen concentration was not determined. Moreover, the composition was found to be homogeneous within each grain of Y_2MgRuO_6 .

Powder X-ray diffraction measurements of a crushed crystal reveal the appearance of Y_2O_3 and MgO impurity phases with a volume fraction of 22.5(1)% and 7.1(3)% respectively, see Figure 2. Nonetheless, the $\sim 70\%$ majority phase of Y_2MgRuO_6 could be refined properly. The obtained lattice constants and structural parameters are listed in Table 1. A refinement of the occupancies of Ru and Mg at the two B-sites reveals a composition of $\text{Y}_2\text{Mg}_{0.902(4)}\text{Ru}_{1.098(4)}\text{O}_6$ which is close to the composition of $\text{Y}_{2.00(1)}\text{Mg}_{0.85(1)}\text{Ru}_{1.11(1)}\text{O}_6$ indicated by the WDX measurements.

For an accurate structural analysis, two single crystals with dimensions of the order of $10\ \mu\text{m}$ were prepared from the original as-grown crystal (sample A and B). Single crystal X-ray diffraction scattering intensities are shown in Figure 3a–c. For sample A, 4677 reflections were collected up to $2\Theta_{\text{max}} = 55.8^\circ$ with a redundancy of 9.10 and an internal R value of 4.02%, whereas 32,124 reflections were collected up to $2\Theta_{\text{max}} = 90.3^\circ$ with a redundancy of 17.89 and an internal R value of 4.74% for sample B. Both measured single crystals show the expected monoclinic twinning for monoclinic systems (with a monoclinic angle $\beta = 90.178(1)^\circ$) with twin fractions of close to 50%, see Table 2. The finally obtained crystal structure is that of a double perovskite as shown in Figure 3d.

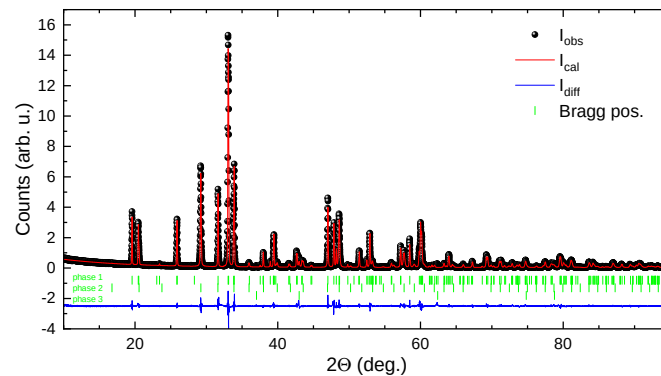


Figure 2. Powder X-ray diffraction pattern of Y_2MgRuO_6 (phase 1) together with impurity phases of Y_2O_3 (phase 2) and MgO (phase 3); $I_{\text{diff}} = I_{\text{obs}} - I_{\text{cal}}$.

Table 1. Refinement results of powder X-ray diffraction measurements of a powdered crystal of Y_2MgRuO_6 with space group $P2_1/n$ (R_p :8.55%, wR_p :12.08%). The obtained composition is $\text{Y}_2\text{Mg}_{0.902(4)}\text{Ru}_{1.098(4)}\text{O}_6$. There was no indication of a Y deficiency. The oxygen content was not refined. For the atomic positions of the light oxygen ions and for the isotropic displacement parameters U_{iso} of all atoms, the results of our single crystal X-ray diffraction measurements (of sample B) were used.

Atom	Wyck.	occ.	x	y	z	U_{iso} ($\text{\AA}^2$)
Ru1	2d	0.972(4)	0	1/2	1/2	0.00346
Y1	4e	1	0.9786(3)	0.92820(17)	0.24838(17)	0.007393
Mg1	2c	0.874(3)	1/2	0	1/2	0.005096
O1	4e	1	0.1872	0.8066	0.5596	0.0077
O2	4e	1	0.1872	0.8063	0.9425	0.0071
O3	4e	1	0.1181	0.5444	0.2507	0.007
Mg2	2d	0.028(4)	0	1/2	1/2	0.00346
Ru2	2c	0.126(3)	1/2	0	1/2	0.005096
latt. param. value						
a ($\text{\AA}$)	5.29465(6)					
b ($\text{\AA}$)	5.66404(6)					
c ($\text{\AA}$)	7.6080(1)					
β ($^\circ$)	90.178(1)					

An initial refinement of the occupancies of Ru versus Mg at both of the two B-sites reveals compositions of $\text{Y}_2\text{Mg}_{0.867(4)}\text{Ru}_{1.133(4)}\text{O}_6$ for sample A and $\text{Y}_2\text{Mg}_{0.853(2)}\text{Ru}_{1.147(2)}\text{O}_6$ for sample B. From the composition $\text{Y}_2\text{Mg}_{0.853(2)}\text{Ru}_{1.147(2)}\text{O}_6$ one would nominally expect a Ru 3.74+ oxidation state appearing in this double perovskite compound. But chemically, a Ru^{4+} oxidation state is expected for these growth conditions. In order to obtain the rather stable Ru^{4+} oxidation state, there would have to be nominally a 5% Y-deficiency of -0.1 in $\text{Y}_2\text{Mg}_{0.853(2)}\text{Ru}_{1.147(2)}\text{O}_6$ which would be in agreement with (i) the appearance of an Y_2O_3 impurity phase in the as-grown crystal, (ii) with somewhat enhanced atomic displacement parameters of the Y ions observed in the refinement and, on the other hand, also (iii) with the compact 113-perovskite structure that does not allow for interstitial sites for excess oxygen. Therefore, the Y occupancy is also refined in a subsequent step. Indeed, this refinement yields compositions of $\text{Y}_{1.909(16)}\text{Mg}_{0.941(9)}\text{Ru}_{1.059(9)}\text{O}_6$ (sample A) and $\text{Y}_{1.880(8)}\text{Mg}_{0.953(4)}\text{Ru}_{1.047(4)}\text{O}_6$ (sample B) that exhibit Y deficiencies. (Due to the refinement of the Y occupancy, an improvement of the R-values could be achieved—e.g., for sample B, the weighted R-value R_w decreased from 5.41% to 5.09%. Furthermore, the B/B'-site disorder was also altered—e.g., the Ru1 occupancy decreased from 0.770(2) to 0.709(4) and the Mg1 occupancy increased from 0.623(2) to 0.661(3) for sample B. On the other hand, the other structural parameters x , y , z and the twin fractions remained very

stable). The finally obtained structural parameters and refinement results are listed in Tables 2–4. These finally obtained compositions would be consistent with a Ru^{4+} oxidation state, assuming the presence of oxygen deficiencies δ , i.e., the occupancy of the oxygen ions should be 98.73% and 97.78% for samples A and B, respectively. (Otherwise, there would be a small admixture of Ru^{5+} ions for $\delta = 0$). We did not refine the occupancies of the oxygen ions and set their occupancies to 100% within the refinement since then there would be too many free parameters together with the scale factor. The refined compositions $\text{Y}_{1.909(16)}\text{Mg}_{0.941(9)}\text{Ru}_{1.059(9)}\text{O}_{6-\delta}$ and $\text{Y}_{1.880(8)}\text{Mg}_{0.953(4)}\text{Ru}_{1.047(4)}\text{O}_{6-\delta}$ for the two samples are very roughly consistent with each other. Why the B/B' -site mixing for these two single crystals is larger than for the powder sample is unclear. One reason could be that the two single crystals originate from a less-ordered part of the as-grown boule. Another possibility would be that single crystal X-ray diffraction gives more accurate results.

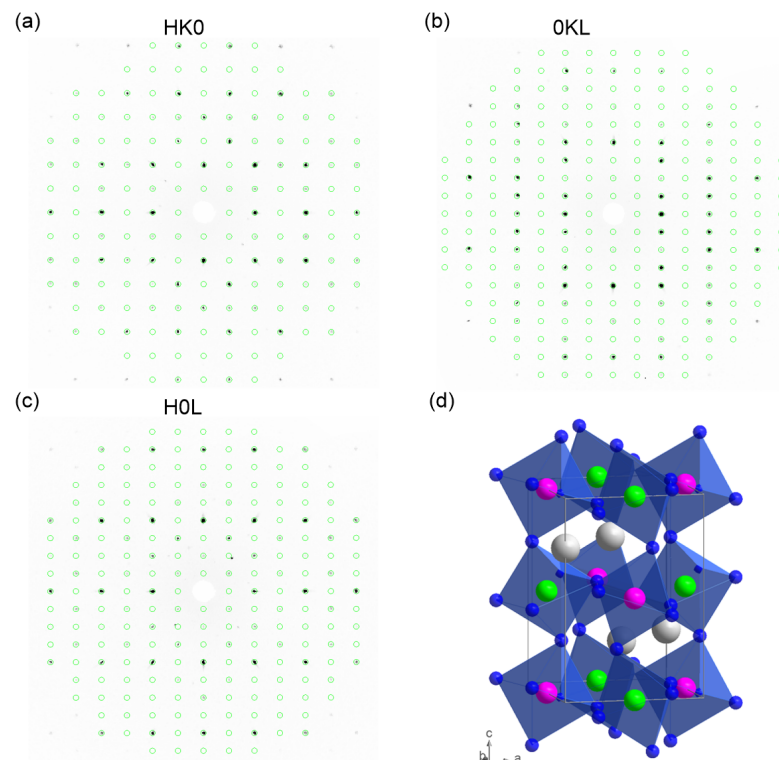


Figure 3. (a–c) X-ray scattering intensities (displayed in a black/white representation) within $HK0$, OKL and $H0L$ planes of reciprocal space (for the measurement of sample B). Green circles indicate the positions for expected Bragg reflections according to space group $P2_1/n$. (d) Crystal structure of Y_2MgRuO_6 ; white/green/magenta/blue spheres: Y/ B/B' /oxygen ions; the B -site is predominantly occupied by Ru and the B' -site, mainly by Mg.

Due to the presence of B/B' -site mixing, the bond valence sum (BVS) formalism seems not very reliable to determine the Ru oxidation state since the so-called Ru–O distances are only a (weighted) average of Ru–O and Mg–O distances due to the B/B' -site disorder. Note that the ionic radii of Mg^{2+} ions is about 0.1 Å larger than that of the Ru ions. And as expected, the formal BVS of Ru ions is lower than for a Ru^{4+} ion, i.e., 3.522(9)+ for $\text{Ru}1^{(B)}$, and correspondingly, these BVS values are somewhat larger than expected for a Mg^{2+} ion, i.e., 2.378(6)+ for $\text{Mg}1^{(B)}$.

Table 2. Crystallographic and structural refinement data of two single crystal X-ray diffraction measurements of Y_2MgRuO_6 (sample A and sample B). The crystallographic software *Jana2006* was used for the structural refinement [23]; *: unobserved reflections ($I < 3\sigma(I)$) were used in the refinement. Note that the lattice parameters determined by single crystal X-ray diffraction are not as reliable as the lattice constants determined by powder X-ray diffraction listed in Table 1 because of poorer resolution and monoclinic twinning.

Crystal	Sample A	Sample B
Temperature	room temperature	room temperature
Wavelength	Mo K_α	Mo K_α
Crystal system	monoclinic	monoclinic
Space group	$P2_1/n$ (14)	$P2_1/n$ (14)
Unit cell dimensions	$a = 5.2834(13) \text{ \AA}$ $b = 5.6571(13) \text{ \AA}$ $c = 7.6134(16) \text{ \AA}$ $\beta = 90.134(11)^\circ$	$a = 5.2885(15) \text{ \AA}$ $b = 5.6670(17) \text{ \AA}$ $c = 7.6114(14) \text{ \AA}$ $\beta = 89.996(2)^\circ$
Volume	$227.55(9) \text{ \AA}^3$	$228.11(10) \text{ \AA}^3$
Z	4	4
$F(000)$	373	373
Crystal size	$\sim 10\text{--}30 \text{ }\mu\text{m}$	$\sim 10\text{--}30 \text{ }\mu\text{m}$
$2\Theta_{\text{max}}$	55.8°	90.3°
Index range	$h: -6 \rightarrow 6$ $k: -7 \rightarrow 7$ $l: -9 \rightarrow 9$	$h: -10 \rightarrow 10$ $k: -11 \rightarrow 11$ $l: -15 \rightarrow 15$
Reflections in total/independent	4677/514	32,124/1796
Observed reflections/independent	4102/476	23,056/1524
Internal R-value	4.02%	4.74%
Completeness up to $2\Theta_{\text{max}}$	98.5%	94%
Completeness up to $2\Theta = 87.15^\circ$		98%
Absorption correction	multi-scan	multi-scan
Min./max. transmission	0.5486/0.7456	0.6431/0.7489
Refinement method	least squares on F^2	least squares on F^2
Reflections threshold *	$I > 3\sigma(I)$	$I > 3\sigma(I)$
Monoclinic twin fraction 1	54.0(3)%	50.7(2)%
Goodness of fit	1.61	1.42
R/ R_w	2.13%/4.99%	2.26%/5.09%

The temperature dependence of magnetic susceptibility $\chi(T)$ for the nominal composition of Y_2MgRuO_6 is shown in Figure 4a. The behavior of $\chi(T)$ resembles that of $\text{La}_2\text{MgRuO}_6$ and $\text{La}_2\text{ZnRuO}_6$ [15,20]. There is no clear evidence for any magnetic order in Y_2MgRuO_6 . In Figure 4a, the inverse magnetic susceptibility is also plotted. As expected for Curie–Weiss behavior, a rather linear regime can be observed above 300 K (as compared to 250 K in $\text{La}_2\text{MgRuO}_6$ and $\text{La}_2\text{ZnRuO}_6$ [15,20]). A Curie–Weiss fit yields an effective moment of $2.67(35) \mu_B$ per formula unit, which is only slightly less than the expected spin-only value for a $S = 1$ system. This result corroborates that the Ru ions are in a $S = 1 \text{ Ru}^{4+}$ oxidation state. Note that $\chi(T)$ is also not temperature independent as would be expected for a $J = 0$ system. Finally, the Weiss-temperature Θ amounts to $27.6(2.8) \text{ K}$, which is indicative for ferromagnetic exchange interactions. This is different from $\text{La}_2\text{MgRuO}_6$ and $\text{La}_2\text{ZnRuO}_6$ where a negative Weiss temperature was observed and might be either related to smaller bond angles due to the small size of the Y ions (the Ru–O–Mg bond angles amount to $143.0(1)^\circ$ in Y_2MgRuO_6) or to a higher degree of B/B' -site disorder in our sample. This significant amount of B/B' -site disorder (observed by precise single crystal X-ray diffraction measurements) is also very likely responsible for the lack of magnetic order and also for the $S = 1$ state due to clustering of Ru at the microscopic level since it leads to a very significant amount of Ru–O–Ru exchange paths instead of the separation of the RuO_6 octahedra by Mg ions in a hypothetical crystal with perfect B/B' -site order.

Isothermal magnetization $M(H)$ measurements of Y_2MgRuO_6 are shown in Figure 4b. At 2 K, the magnetization shows an S-shaped field dependence, which could be explained by the presence of ferromagnetic exchange interactions in this compound, indicated by the positive Weiss temperature.

Table 3. Refinement results of single crystal X-ray diffraction measurements of Y_2MgRuO_6 (sample A). Note that the lattice parameters determined by single crystal X-ray diffraction are not as reliable as the lattice constants determined by powder X-ray diffraction. Therefore, only the values in Table 1 should be used.

Composition: Y _{1.909(16)} Mg _{0.941(9)} Ru _{1.059(9)} O ₆						
atom	occ.	x	y	z	U _{iso} (Å ²)	
Ru1	0.786(10)	0	0.5	0.5	0.0045(2)	
Y1	0.955(8)	0.97912(8)	0.92762(9)	0.24949(9)	0.00884(18)	
Mg1	0.727(6)	0.5	0	0.5	0.0070(4)	
O1	1	0.1852(9)	0.8040(7)	0.5605(5)	0.0110(12)	
O2	1	0.1881(8)	0.8084(7)	0.9424(6)	0.0119(13)	
O3	1	0.1178(6)	0.5453(6)	0.2520(6)	0.0117(10)	
Mg2	0.214(10)	0	0.5	0.5	0.0045(2)	
Ru2	0.273(6)	0.5	0	0.5	0.0070(4)	
atom	U ₁₁ (Å ²)	U ₂₂ (Å ²)	U ₃₃ (Å ²)	U ₁₂ (Å ²)	U ₁₃ (Å ²)	U ₂₃ (Å ²)
Ru1	0.0045(3)	0.0044(4)	0.0045(3)	0.0007(14)	0.0026(8)	0.0008(9)
Y1	0.0089(3)	0.0079(3)	0.0098(3)	0.00059(17)	−0.0011(13)	−0.00014(15)
Mg1	0.0066(7)	0.0072(8)	0.0071(7)	0.002(3)	0.003(3)	−0.0006(18)
O1	0.012(2)	0.0095(18)	0.012(2)	−0.0006(19)	0.000(2)	−0.0019(15)
O2	0.012(2)	0.0083(18)	0.015(3)	−0.0026(19)	−0.001(2)	0.0002(15)
O3	0.0124(16)	0.0126(18)	0.0102(18)	−0.0008(13)	0.002(3)	−0.0006(17)
Mg2	0.0045(3)	0.0044(4)	0.0045(3)	0.0007(14)	0.0026(8)	0.0008(9)
Ru2	0.0066(7)	0.0072(8)	0.0071(7)	0.002(3)	0.003(3)	−0.0006(18)

Table 4. Refinement results of single crystal X-ray diffraction measurements of Y_2MgRuO_6 (sample B). Note that the lattice parameters determined by single crystal X-ray diffraction are not as reliable as the lattice constants determined by powder X-ray diffraction. Therefore, only the values in Table 1 should be used.

Composition: Y _{1.880(8)} Mg _{0.953(4)} Ru _{1.047(4)} O ₆						
atom	occ.	x	y	z	U _{iso} (Å ²)	
Ru1	0.709(4)	0	0.5	0.5	0.00327(6)	
Y1	0.940(4)	0.97899(4)	0.92746(4)	0.24962(5)	0.00717(5)	
Mg1	0.661(3)	0.5	0	0.5	0.00485(12)	
O1	1	0.1865(5)	0.8066(4)	0.5595(3)	0.0100(5)	
O2	1	0.1880(4)	0.8063(4)	0.9425(3)	0.0088(5)	
O3	1	0.1180(3)	0.5444(3)	0.2503(3)	0.0088(3)	
Mg2	0.291(4)	0	0.5	0.5	0.00327(6)	
Ru2	0.339(3)	0.5	0	0.5	0.00485(12)	
atom	U ₁₁ (Å ²)	U ₂₂ (Å ²)	U ₃₃ (Å ²)	U ₁₂ (Å ²)	U ₁₃ (Å ²)	U ₂₃ (Å ²)
Ru1	0.00340(11)	0.00314(11)	0.00326(11)	0.0014(3)	0.0021(2)	−0.0009(3)
Y1	0.00755(8)	0.00573(9)	0.00823(9)	0.00083(6)	−0.0017(4)	−0.00001(8)
Mg1	0.0052(2)	0.0051(2)	0.00433(19)	0.0028(5)	0.0025(6)	0.0023(5)
O1	0.0119(11)	0.0078(8)	0.0104(9)	−0.0007(8)	−0.0014(9)	−0.0014(7)
O2	0.0072(9)	0.0091(8)	0.0102(9)	−0.0024(8)	−0.0031(8)	0.0021(7)
O3	0.0105(5)	0.0094(6)	0.0066(5)	0.0007(4)	−0.0008(10)	−0.0018(7)
Mg2	0.00340(11)	0.00314(11)	0.00326(11)	0.0014(3)	0.0021(2)	−0.0009(3)
Ru2	0.0052(2)	0.0051(2)	0.00433(19)	0.0028(5)	0.0025(6)	0.0023(5)

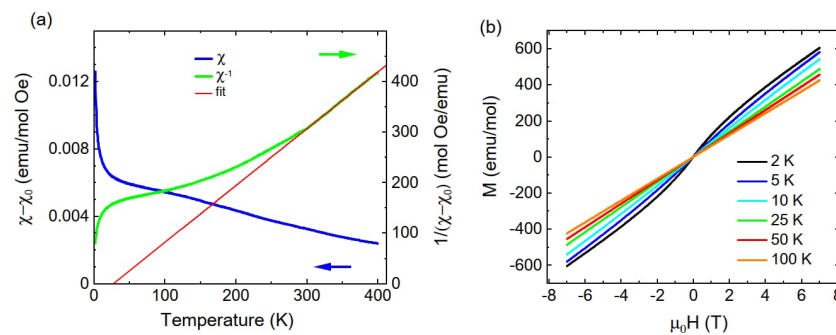


Figure 4. (a) The temperature dependence of magnetic susceptibility and inverse susceptibility of Y_2MgRuO_6 (χ_0 was obtained from the Curie–Weiss fit, and $\chi - \chi_0$ is plotted in order to visualize the linear behavior of the corresponding inverse susceptibility between 300 K and 400 K). (b) Magnetization $M(H)$ measured at different temperatures of Y_2MgRuO_6 .

3. Materials and Methods

Y_2MgRuO_6 crystals were grown in an optical floating zone furnace (HKZ from Scientific Instruments Dresden GmbH, Dresden, Germany) equipped with a Xe lamp. The synthesis temperature was of the order of $\approx 1800^\circ\text{C}$. As starting materials, RuO_2 (Aldrich 99.9%), Y_2O_3 (Alfa Aesar 99.9%) and MgO (Alfa Aesar 99.95%) were used.

Powder X-ray diffraction (XRD) measurements were performed on a powder X-ray diffractometer (Bruker D8 Discover A25, Bruker AXS GmbH, Karlsruhe, Germany) ($\text{Cu } K_{\alpha 1}$ radiation) diffractometer at room temperature.

Single crystal X-ray diffraction (XRD) measurements were performed on a single crystal X-ray diffractometer (Bruker D8, Bruker AXS GmbH, Karlsruhe, Germany) with a bent graphite monochromator for about 3 times the intensity enhancement that is equipped with a Photon III CMOS area detector ($\text{Mo } K_{\alpha}$ radiation).

The magnetization and susceptibility measurements were performed using a *Magnetic Property Measuring System (MPMS)*, Quantum Design, San Diego, CA, USA.

Wavelength dispersive X-ray analysis was performed on an electron microprobe (Cameca, SX100, Munich, Germany). Electron backscatter diffraction (EBSD) measurements were realized on a scanning electron microscope (JSM-7800F, Jeol (Germany GmbH), Munich, Germany) equipped with an EBSD system (CrystAlign, Bruker Nano GmbH, Berlin, Germany).

4. Conclusions

Single crystals of a new double perovskite— Y_2MgRuO_6 —could be synthesized with the floating zone technique. The fact that the solid state synthesis of powder samples of the nominal composition Y_2MgRuO_6 entirely failed might indicate that the synthesis conditions (c,p,T) reached within the growth chamber could be important for the successful growth of this phase. Due to inclusions of Y_2O_3 and MgO impurity phases, only crystal sizes of up to $\sim 40\ \mu\text{m}$ could be prepared from the as-grown boule. Powder and single crystal X-ray diffraction measurements indicate a monoclinic crystal structure with space group $P2_1/n$ and a monoclinic angle $\beta = 90.178(1)^\circ$. In addition, these measurements indicate a small excess of Ru and non-negligible B/B' -site mixing in this compound. Single crystal X-ray diffraction measurements reveal a composition of $\text{Y}_{1.880(8)}\text{Mg}_{0.953(4)}\text{Ru}_{1.047(4)}\text{O}_{6-\delta}$ for one of these crystals. The magnetic susceptibility of this double perovskite resembles that of $\text{La}_2\text{MgRuO}_6$ or $\text{La}_2\text{ZnRuO}_6$. There is no clear indication for any long-range magnetic order in this material. Also, no Van Vleck-type temperature-independent magnetism—as expected for a $J = 0$ system—can be observed in this compound. Instead, the effective moment obtained from a Curie–Weiss fit is indicative for a $S = 1\ \text{Ru}^{4+}$ state. This could be probably the consequence of the B/B' -site disorder, which induces a large amount of Ru–O–Ru exchange paths like in single perovskites as, for example, SrRuO_3 . In contrast

to $\text{La}_2\text{MgRuO}_6$ or $\text{La}_2\text{ZnRuO}_6$, the Weiss constant Θ is positive, indicating ferromagnetic exchange interactions in Y_2MgRuO_6 .

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References

1. Khaliullin, G. Excitonic Magnetism in Van Vleck-type d^4 Mott Insulators. *Phys. Rev. Lett.* **2013**, *111*, 197201. [\[CrossRef\]](#)
2. Meetei, O.N.; Cole, W.S.; Randeria, M.; Trivedi, N. Novel magnetic state in d^4 Mott insulators. *Phys. Rev. B* **2015**, *91*, 054412. [\[CrossRef\]](#)
3. Takayama, T.; Chaloupka, J.; Smerald, A.; Khaliullin, G.; Takagi, H. Spin–Orbit-Entangled Electronic Phases in 4d and 5d Transition-Metal Compounds. *J. Phys. Soc. Jpn.* **2021**, *90*, 062001. [\[CrossRef\]](#)
4. Takahashi, H.; Suzuki, H.; Bertinshaw, J.; Bette, S.; Mühle, C.; Nuss, J.; Dinnebier, R.; Yaresko, A.; Khaliullin, G.; Gretarsson, H.; et al. Nonmagnetic $J = 0$ State and Spin-Orbit Excitations in K_2RuCl_6 . *Phys. Rev. Lett.* **2021**, *127*, 227201. [\[CrossRef\]](#)
5. Chaloupka, J.; Khaliullin, G. Highly frustrated magnetism in relativistic d^4 Mott insulators: Bosonic analog of the Kitaev honeycomb model. *Phys. Rev. B* **2019**, *100*, 224413. [\[CrossRef\]](#)
6. Anisimov, P.S.; Aust, F.; Khaliullin, G.; Daghofer, M. Nontrivial Triplon Topology and Triplon Liquid in Kitaev-Heisenberg-type Excitonic Magnets. *Phys. Rev. Lett.* **2019**, *122*, 177201. [\[CrossRef\]](#)
7. Kaushal, N.; Soni, R.; Nocera, A.; Alvarez, G.; Dagotto, E. BCS-BEC crossover in a $(t_2g)^4$ excitonic magnet. *Phys. Rev. B* **2020**, *101*, 245147. [\[CrossRef\]](#)
8. Khalifah, P.; Osborn, R.; Huang, Q.; Zandbergen, H.W.; Jin, R.; Liu, Y.; Mandrus, D.; Cava, R.J. Orbital Ordering Transition in $\text{La}_4\text{Ru}_2\text{O}_{10}$. *Science* **2002**, *297*, 2237. [\[CrossRef\]](#)
9. Wu, H.; Hu, Z.; Burnus, T.; Denlinger, J.D.; Khalifah, P.G.; Mandrus, D.G.; Jang, L.Y.; Hsieh, H.H.; Tanaka, A.; Liang, K.S.; et al. Orbitally Driven Spin-Singlet Dimerization in $S = 1$ $\text{La}_4\text{Ru}_2\text{O}_{10}$. *Phys. Rev. Lett.* **2006**, *96*, 256402. [\[CrossRef\]](#)
10. Jain, J.; Krautloher, M.; Porras, J.; Ryu, G.H.; Chen, D.P.; Abernathy, D.L.; Park, J.T.; Ivanov, A.; Chaloupka, J.; Khaliullin, G.; et al. Higgs mode and its decay in a two-dimensional antiferromagnet. *Nat. Phys.* **2017**, *13*, 163. [\[CrossRef\]](#)
11. Miura, Z.; Sato, M.; Yamakawa, Y.; Habaguchi, T.; Ōno, Y. Structural Transition of Li_2RuO_3 Induced by Molecular-Orbit Formation. *J. Phys. Soc. Jpn.* **2009**, *78*, 094706. [\[CrossRef\]](#)
12. Kumar, R.; Dey, T.; Ette, P.M.; Ramesha, K.; Chakraborty, A.; Dasgupta, I.; Orain, J.C.; Baines, C.; Tóth, S.; Shahee, A.; et al. Unconventional magnetism in the $4d^4$ -based $S = 1$ honeycomb system $\text{Ag}_3\text{LiRu}_2\text{O}_6$. *Phys. Rev. B* **2019**, *99*, 054417. [\[CrossRef\]](#)
13. Takayama, T.; Blankenhorn, M.; Bertinshaw, J.; Haskel, D.; Bogdanov, N.A.; Kitagawa, K.; Yaresko, A.N.; Krajewska, A.; Bette, S.; McNally, G.; et al. Competing spin-orbital singlet states in the $4d^4$ honeycomb ruthenate $\text{Ag}_3\text{LiRu}_2\text{O}_6$. *Phys. Rev. Res.* **2022**, *4*, 043079. [\[CrossRef\]](#)
14. Kimchi, I.; Vishwanath, A. Kitaev-Heisenberg models for iridates on the triangular, hyperkagome, kagome, fcc, and pyrochlore lattices. *Phys. Rev. B* **2014**, *89*, 014414. [\[CrossRef\]](#)
15. Dass, R.I.; Yan, J.Q.; Goodenough, J.B. Ruthenium double perovskites: Transport and magnetic properties. *Phys. Rev. B* **2004**, *69*, 094416. [\[CrossRef\]](#)
16. Battle, P.D.; Frost, J.R.; Kim, S.H. Magnetic Properties of $\text{La}_2\text{CuTi}_{1-x}\text{Ru}_x\text{O}_6$ and LaZnRuO_6 . *J. Mater. Chem.* **1995**, *5*, 1003. [\[CrossRef\]](#)
17. Choy, J.H.; Kim, J.Y.; Hwang, S.H.; Kim, S.J.; Demazeau, G. Soft XAFS study on the 4d electronic structure of ruthenium in complex perovskite oxide. *Int. J. Inorg. Mater.* **2000**, *2*, 61. [\[CrossRef\]](#)
18. Seinen, P.; van Berkel, F.; Groen, W.; Ijdo, D. The ordered perovskite system $\text{Ln}_2\text{NiRuO}_6$. *Mater. Res. Bull.* **1987**, *22*, 535. [\[CrossRef\]](#)
19. Battle, P.; Jones, C. The crystal structure of $\text{Ln}_2\text{NiRuO}_6$. *Mater. Res. Bull.* **1987**, *22*, 1623. [\[CrossRef\]](#)
20. Yoshii, K.; Ikeda, N.; Mizumaki, M. Magnetic and dielectric properties of the ruthenium double perovskites La_2MRuO_6 ($M = \text{Mg}, \text{Co}, \text{Ni}$, and Zn). *Phys. Stat. Sol.* **2008**, *203*, 2812. [\[CrossRef\]](#)
21. Li, Z.; Liu, C.F.; Skoulatos, M.; Tjeng, L.; Komarek, A. Floating zone growth of Ba-substituted ruthenate $\text{Sr}_{2-x}\text{Ba}_x\text{RuO}_4$. *J. Cryst. Growth* **2015**, *427*, 94. [\[CrossRef\]](#)

22. Kennedy, B.J. Structure refinement of $\text{Y}_2\text{Ru}_2\text{O}_7$ by neutron powder diffraction. *Acta Cryst.* **1995**, *C51*, 790–792. [[CrossRef](#)]
23. Petricek, V.; Dusek, M.; Palatinus, L. *Z. Kristallogr.* **2014**, *229*, 345.

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