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Influence of Hydrostatic Pressure on Magnetic Properties of (Sr,La)(Ru,Cr,Mn)O₃

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An influence of hydrostatic pressure, P , on phase transition temperature, T_C , and spontaneous magnetisation, M_0 , of selected perovskite ruthenates (SrRuO₃, La_{0.2}Sr_{0.8}RuO₃, SrRu_{0.9}Mn_{0.1}O₃, and SrRu_{0.9}Cr_{0.1}O₃) was determined to 12 kbar. A decrease in T_C with pressure was found for all of the studied samples. The $M_0(P)$ remains unchanged for most of the samples, except for the La_{0.2}Sr_{0.8}RuO₃ sample. The weakening of ferromagnetic interactions with increasing pressure is consistent with complex band structure effects related to the modulation of the Ru–O hybridisation by the change of structural distortion.

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1. Introduction

SrRuO₃, the ABO₃-perovskite, is known as the only 4*d* transition metal oxide that exhibits clear ferromagnetic (FM) properties. The ferromagnetism in SrRuO₃, with T_C about 160 K [1], arises from a parallel alignment of magnetic moments of 4*d* t_{2g} electrons leading to the low-spin electronic configuration of Ru⁴⁺ ions [2, 3]. Strong Ru t_{2g} –O $2p$ hybridisation leads to an itinerant character of ferromagnetism in this compound, evidenced by metallic conductivity, reduced magnetic moment, and lack of saturation of magnetisation in dc field of 300 kOe [3, 4]. Different substitutions in SrRuO₃, namely, by lanthanide ions or alkaline earth ions at the A site [4–7] and 3*d* transition metal ions at the B site [8], result in rapid changes of magnetic properties. The heterovalent La³⁺ substitution for Sr²⁺ causes a pronounced suppression of ferromagnetism by attenuation of Ru t_{2g} –O $2p$ hybridisation brought by the valence change of Ru and leads to antiferromagnetism

in LaRuO₃ [9]. The T_C decreases to about 75 K for La_{0.2}Sr_{0.8}RuO₃ [7]. The substitution of Ru ion by Mn⁴⁺ (d^3) eliminates one of the Ru t_{2g} electrons, weakens the itinerant character of the d electrons, and introduces antiferromagnetic (AFM) interactions. As a result ferromagnetism is rapidly suppressed to $T_C = 121$ K for SrRu_{0.9}Mn_{0.1}O₃ [10]. The Cr-substitution leads to unique enhancement of T_C up to 188 K [8] for SrRu_{0.9}Cr_{0.1}O₃ due to possibility of electron transfer from Ru⁴⁺ to Cr⁴⁺ (d^2) leading to minority-band double exchange (DE) interaction [11].

The external pressure is a useful tool to tune the magnetism in these oxides by compression of the A–O and B–O bonds that may influence the structural distortion and consequently, the Ru–O hybridisation as postulated by Mazin and Singh [3]. Neumeier et al. studied the pressure effect on magnetic and transport properties in perovskite ruthenate [12], showing that the value of pressure coefficient dT_C/dP , equal to -0.57 K/kbar, is in qualitative agreement with the predictions of the Wohlfarth model.

In this paper, we present the results of magnetic measurements performed for several substituted perovskite ruthenates under hydrostatic pressure to 12 kbar.

2. Experimental details

The stoichiometric SrRuO₃, La_{0.2}Sr_{0.8}RuO₃, and the SrRu_{0.9}M_{0.1}O₃ (M = Mn and Cr) samples have been prepared using standard ceramic synthesis method [13]. All the magnetic measurements were performed in the temperature range of 5–250 K at magnetic field up to 16 kOe using a PAR 4500 vibrating sample magnetometer. A temperature dependence of magnetisation was measured using zero-field-cooling (ZFC) and field-cooling (FC) procedure, whereas $M(H)$ dependence was measured after FC in maximum applied field. For these measurements a miniature container of CuBe [14] with an inside diameter of 1.42 mm was employed as a pressure cell. A mixture of mineral oil–kerosene was used as a pressure transmitting medium. The pressure at low temperature was determined by the pressure dependence of the superconducting transition temperature of pure tin sensor placed near the sample.

3. Results and discussion

From the ZFC and FC magnetisation curves (Figs. 1a, 2a, 3a, 4a) it can be seen that the ferromagnetic transition temperature T_C decreases with pressure for all samples. T_C was determined from the magnetisation $M(T)$ curves as the temperature of the maximum slope of the derivative of $M(T)$, $-dM/dT$. In Figs. 1b, 2b, 3b, 4b hysteresis loops measured at $T = 10$ K are shown. All samples show the clear spontaneous FM moment M_0 , which was extracted by linear extrapolation of $M(H)$ from a high-field region to $H = 0$. The existence of M_0 together with a marked divergence between M_{ZFC} and M_{FC} at low temperatures confirms robust ferromagnetism below T_C reported earlier for these compounds [2, 7, 8]. The M_0 does not change under pressure for most of the samples, except La_{0.2}Sr_{0.8}RuO₃,

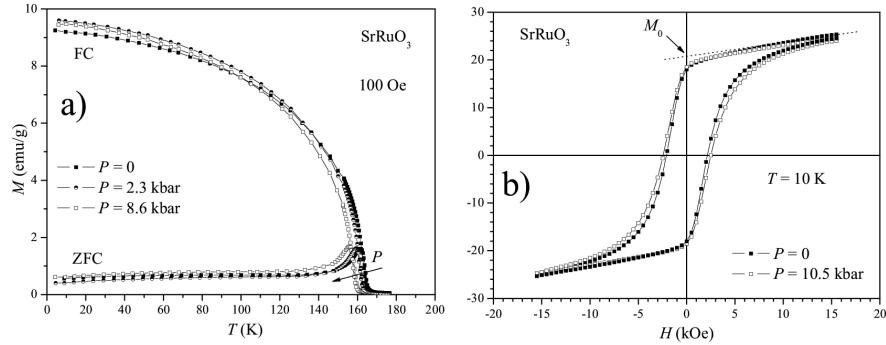


Fig. 1. (a) Temperature dependence of M_{ZFC} and M_{FC} for SrRuO_3 measured at 100 Oe. (b) Magnetisation hysteresis loops for SrRuO_3 at $T = 10$ K.

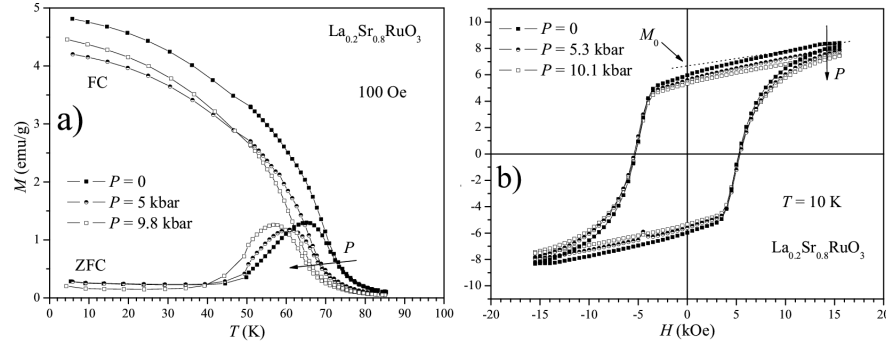


Fig. 2. (a) Temperature dependence of M_{ZFC} and M_{FC} for $\text{La}_{0.2}\text{Sr}_{0.8}\text{RuO}_3$ measured at 100 Oe. (b) Magnetisation hysteresis loops for $\text{La}_{0.2}\text{Sr}_{0.8}\text{RuO}_3$ at $T = 10$ K.

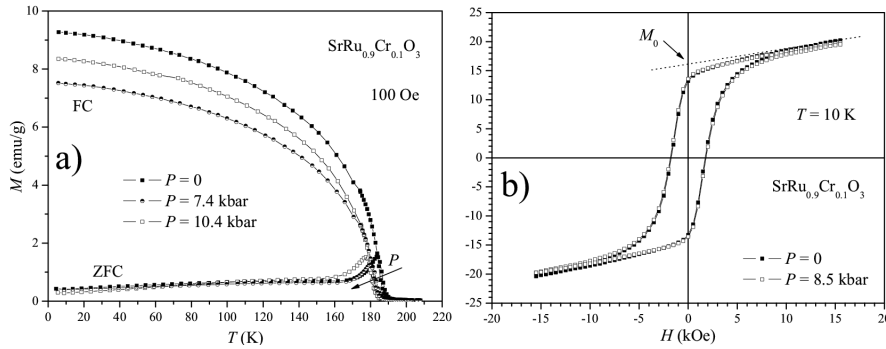


Fig. 3. (a) Temperature dependence of M_{ZFC} and M_{FC} for $\text{SrRu}_{0.9}\text{Cr}_{0.1}\text{O}_3$ measured at 100 Oe. (b) Magnetisation hysteresis loops for $\text{SrRu}_{0.9}\text{Cr}_{0.1}\text{O}_3$ at $T = 10$ K.

where it decreases with increasing pressure. The observed changes of T_C and M_0 can be approximated by linear pressure dependence with the pressure coefficients of dT_C/dP and dM_0/dP for T_C and M_0 , respectively.

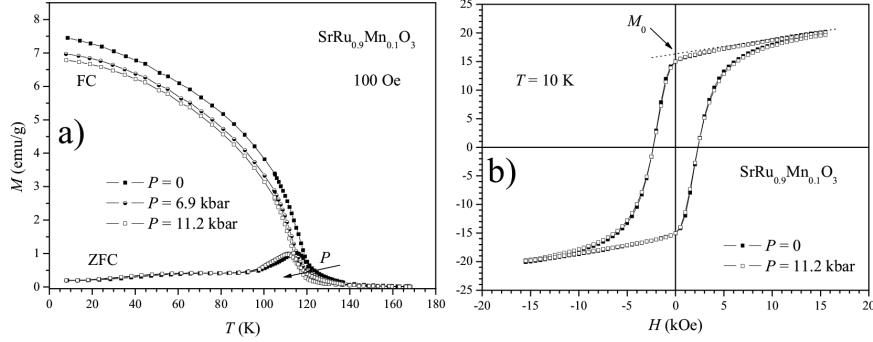


Fig. 4. (a) Temperature dependence of M_{ZFC} and M_{FC} for $\text{SrRu}_{0.9}\text{Mn}_{0.1}\text{O}_3$ measured at 100 Oe. (b) Magnetisation hysteresis loops for $\text{SrRu}_{0.9}\text{Mn}_{0.1}\text{O}_3$ at $T = 10$ K.

The decrease in T_C with pressure for SrRuO_3 was reported earlier and explained in terms of compressive strain acting on sample [11]. Our pressure coefficient $dT_C/dP = -0.68$ K/kbar is in good agreement with that reported in Ref. [11] (-0.57 K/kbar). A considerable suppression of T_C in $\text{La}_{0.2}\text{Sr}_{0.8}\text{RuO}_3$, when compared to that of SrRuO_3 at ambient pressure has been attributed to attenuation of Ru–O hybridisation, caused by chemical pressure from La ion [7]. The external pressure creates further attenuation of Ru–O hybridisation evidenced by negative pressure coefficient (-0.67 K/kbar), in such a way that some magnetic moments become more localised and as a result the $\text{Ru}_{t2g}\text{--O}_{2p}\text{--Ru}_{t2g}$ AFM superexchange interactions are expected to develop at the expense of FM ones [7]. This may also cause a decrease in M_0 with pressure ($dM_0/dP = -0.07$ emu/(g kbar)) for this sample. In the case of $\text{SrRu}_{0.9}\text{Cr}_{0.1}\text{O}_3$ we observe a weakening of $\text{Ru}_{t2g}\text{--O}_{2p}\text{--Cr}_{t2g}$ DE interactions evidenced by a decrease in T_C with increasing pressure ($dT_C/dP = -0.53$ K/kbar). The spin-down electron in Ru_{t2g} band becomes more localised giving no contribution to AFM coupling of the Ru and Cr ions [10]. Therefore, M_0 remains constant with increasing pressure. For $\text{SrRu}_{0.9}\text{Mn}_{0.1}\text{O}_3$ the external pressure probably affects Ru–O hybridisation in such a way that it limits the itinerancy of the Ru 4d electrons and consequently weakens the FM coupling. It is evidenced by the decrease in T_C with increasing pressure ($dT_C/dP = -0.39$ K/kbar). The M_0 remains constant with increasing pressure, similar to that of SrRuO_3 and $\text{SrRu}_{0.9}\text{Cr}_{0.1}\text{O}_3$. For all these compounds the FM phase volume does not change under pressure. Thus, unlike substitutions on the A-site, substitutions for Ru seem to be affected in such a way that only T_C 's are changed [9].

4. Conclusions

An influence of hydrostatic pressure on phase transition temperature, T_C , and spontaneous magnetisation, M_0 , of $(\text{Sr},\text{La})(\text{Ru},\text{Cr},\text{Mn})\text{O}_3$ was investigated. We observed a decrease in T_C with pressure for all of the samples independent of

the sign of charge doping or its affect on the FM transition. The $M_0(P)$ remains unchanged for most of the samples, except for the $\text{La}_{0.2}\text{Sr}_{0.8}\text{RuO}_3$ sample, where M_0 decreases with pressure. The weakening of FM interactions with increasing pressure should thus be associated with a general feature of the band structure that is related to the modulation of the Ru–O hybridisation by the change of structural distortion.

Acknowledgments

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References

- [1] A. Callaghan, C.W. Moeller, R. Ward, *Inorg. Chem.* **5**, 1572 (1966).
- [2] L. Klein, J.S. Dodge, C.H. Ahn, J.W. Reiner, L. Mieville, T.H. Geballe, M.R. Beasley, A. Kapitulnik, *J. Phys., Condens. Matter* **8**, 10111 (1996).
- [3] I.I. Mazin, D.J. Singh, *Phys. Rev. B* **56**, 2556 (1997).
- [4] G. Cao, S. McCall, M. Shepard, J.E. Crow, R.P. Guertin, *Phys. Rev. B* **56**, 321 (1997).
- [5] A. Kanbayasi, *J. Phys. Soc. Jpn.* **44**, 108 (1978).
- [6] M.F. Da Costa, R. Greatrex, N.N. Greenwood, *J. Solid State Chem.* **20**, 381 (1977).
- [7] H. Nakatsugawa, E. Iguchi, Y. Oohara, *J. Phys., Condens. Matter* **14**, 415 (2002).
- [8] L. Pi, A. Maignan, R. Retoux, B. Raveau, *J. Phys., Condens. Matter* **14**, 7391 (2002).
- [9] R.J. Bouchard, J.F. Weiher, *J. Solid State Chem.* **4**, 80 (1972).
- [10] G. Cao, S. Chikara, X.N. Lin, E. Elhami, V. Durairaj, P. Schlottmann, *Phys. Rev. B* **71**, 035104 (2005).
- [11] B. Dabrowski, S. Kolesnik, O. Chmaissem, T. Maxwell, M. Avdeev, P.W. Barnes, J.D. Jorgensen, *Phys. Rev. B* **72**, 054428 (2005).
- [12] J.J. Neumeier, A.L. Cornelius, J.S. Schilling, *Physica B* **198**, 324 (1994).
- [13] B. Dabrowski, O. Chmaissem, P.W. Klamut, S. Kolesnik, M. Maxwell, J. Mais, Y. Ito, B.D. Armstrong, J.D. Jorgensen, S. Short, *Phys. Rev. B* **70**, 014423 (2004).
- [14] M. Baran, V. Dyakonov, L. Gladczuk, G. Levchenko, S. Piechota, H. Szymczak, *Physica C* **241**, 383 (1995).