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Investigation of Possible Half-Metallic Antiferromagnets on Double Perovskites $A_2BB'O_6$ (A=Ca, Sr Ba; B,B'=Transition Metal Elements)

S. H. Chen[1,*], Z. R. Xiao[2], Y. P. Liu[3] and Y. K. Wang[4,*]

[1] *Institute of Physics, Academia Sinica, Taipei 11529, Taiwan.*
[2] *Graduate Institute of Applied Physics, National Chengchi University, Taipei 11605, Taiwan.*
[3] *Department of Physics, National Taiwan Normal University, Taipei 106, Taiwan.*
[4] *Center for General Education and Department of Physics, National Taiwan Normal University, Taipei 106, Taiwan.*

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Abstract. A search was made for possible half-metallic (HM) antiferromagnet (AFM) in all the (C_2^{29}=406) double perovskites structures of $Sr_2BB'O_6$ where BB' pairs are any combination of 3d, 4d or 5d transition elements with the exception of La. Sr can also be replaced by Ca or Ba whenever HM-AFM was found and similar calculations were then performed in order to probe further possibilities. It was found that A_2MoOsO_6, A_2TcReO_6, A_2CrRuO_6, where A=Ca, Sr, Ba, are all potential candidates for HM-AFM. The AFM of $A_2BB'O_6$ comes from both the superexchange mechanism and the generalized double exchange mechanism via the $B(t_{2g})$-O2p_π-$B'(t_{2g})$ coupling, With the latter also being the origin of their HM. Also considered were the effects of spin-orbit coupling (SOC) and correlation (+U) by introducing +SOC and +U corrections. It is found that the SOC effect has much less influence than the correlation effect on the HM property of the compounds. For A_2TcReO_6 and A_2CrRuO_6, after +U, they become nearly Mott-Insulators. In the future, it is hoped that there will be further experimental confirmation for these possible HM-AFM candidates.

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Key words: Half-metal antiferromagnet, GGA +U, superexchange, generalized double exchange.

*Corresponding author. *Email addresses:* chen_shao_hua197@yahoo.com.tw (S. H. Chen), zrxiao@gmail.com (Z. R. Xiao), viva.guitarra@gmail.com (Y. P. Liu), kant@ntnu.edu.tw (Y. K. Wang)

1 Introduction

The characteristic properties of half-metallic (HM) materials are spin magnetic moments quantized with a fully spin-polarized state at the Fermi level, as well as zero spin susceptibility. In 1983, R. de Groot et al. [1] discovered HM ferromagnets (FM) by calculating the band structure of the magnetic semi-Heusler compounds NiReSb and PtReSb. Due to their single-spin charge carriers, HM materials can be applied as single-spin electron sources and high-efficiency magnetic sensors [2–11]. If the total magnetic moment is zero in HM material, it is called HM antiferromagnet (AFM). The first HM-AFM was proposed by van Leuken and de Groot [12] in 1995. HM-AFMs have the following properties: First, they do not carry any macroscopic magnetic field in zero temperature, even thought small magnetization might be induced due to finite temperature spin fluctuations [13]. Second, they can transport a 100% spin polarized charge without net magnetization. Third, theirs magnetic susceptibility is zero. Based on these features, HM-AFMs have several applications: they can be used as probes in spin-polarized scanning tunneling microscopes since they will not disturb the spin character of the sample; as well they play an important role in spintronic devices. However, up till now, there still exists no experimental verification for them.

Many structures are considered as possible HM-AFM candidates, e.g. half-Heusler [12], full-Heusler alloys [14–17], thiospinels [18], Fe-based superconductor [19] and superlattices [20, 21]. They can also be found in some disorder systems, e.g. vacancy-induced rock salt transition metal oxides [23], Co-substituted Heusler alloys [24], and diluted antiferromagnetic (AFM) semiconductors [25, 26]. Pickett [27] first looked for HM-AFMs using the (fixed) cubic double perovskite structure $La_2M'M''O_6$ in 1998. He proposed La_2VMnO_6 and La_2VCuO_6 as two promising candidates for HM-AFM. He also mentioned that the perovskite crystal structure AMO_3, due to its simple crystal structure, potentially large number of members, and strong coupling between magnetic ordering and electronic properties, appeared to be an ideal system in the search for HM-AFM members.

Based on a systematic first-principle study of the ordered double perovskites $A_2BB'O_6$, whose nominal ionic picture is $A_2^{2+}(BB')^{8+}O_6^{2-}$, with possible B and B' pairs in the $3d$, $4d$ and $5d$ transition metal elements. We predict A_2CrRuO_6, A_2MoOsO_6 and A_2TcReO_6 as possible HM-AFM candidates. Among them, only Sr_2CrRuO_6 was previously predicted by Lee and Pickett [32]. We considered two stacked structures ([111], space group $Fm\bar{3}m$, and [001], space group $P4/mmm$, as seen in Fig. 1) with two magnetic states (FM and nonconventional AF (labeled as AF-I), whose spin moment of (B, B, B', B') is (+,+,−,−,)). Furthermore, we also considered the possibilities of a third magnetic states: conventional AF (labeled as AF-II, whose spin moment of (B, B, B', B') is (+,−,+,−,)), which has seldom been compared with AF-I in term of stability in theoretical predictions of HM-AFMs in double perovskites structures.

For transition metal oxides, we took into account spin-orbit coupling (SOC) and correlation ($+U$) effects. The Hubbard U values for all transition metals are reported [33]