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The half-metallic ferromagnetic characters of (001)-oriented thin films of the double perovskite Pb_2FeMoO_6

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ABSTRACT

The structural, electronic and magnetic properties of the three possible terminations of the (001)-oriented thin film of the double perovskite Pb_2FeMoO_6, 10-L $FeMoO_4$ and PbO terminated, 9-L $FeMoO_4$ terminated and 11-L PbO terminated, have been studied by using the first-principles calculations. It is found that, firstly an outwards relaxation is observed for several layers near surface and the relaxed fractional rumpling *s* of the PbO layer is larger than that of the adjacent $FeMoO_4$ layer and both have a decrease tend from surface layer to inner layer. Second, whether parallel to or perpendicular to the surface, the Mo—O bond length is always shorter than the adjacent Fe—O bond length. Thirdly, the half-metallic ferromagnetic (HM-FM) character ensures these three terminations of the (001)-oriented thin films of the double perovskite Pb_2FeMoO_6 a potential application in magnetoresistive and spintronics devices. Fourth, delocalized distribution of the spin charge densities around the Fe atoms on the first $FeMoO_4$ layer leads to a smaller magnetic moment of the Fe atoms on the first $FeMoO_4$ layer than the inner Fe atoms.

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1. Introduction

Double perovskites, firstly discovered by Ward et al. in 1961 [1,2], are a broad class of compounds with a general chemical formula of $A_2BB'O_6$ [3]. Where A is usually an alkaline-earth metal atom (Ca, Sr, Ba) or a rare-earth metal atom (La, Ce, Nd), B a 3*d* (Cr, Mn, Fe, Co, Ni, Zn) and B′ a 4*d* (Mo, Te, Ru) or 5*d* (W, Re, Os) transition-metal (TM) atom. A recent finding of the intrinsic tunneling magnetoresistance (TMR) effect at room temperature in Sr_2FeMoO_6 [4] revives the extensive investigates on the double perovskites. The band structures reveal that they are half-metallic ferromagnetic (HM-FM) with completely (100%) spin-polarized transport properties at the Fermi level [3,4]. The Curie temperature T_C of the Sr_2FeMoO_6 is found to be 415 K much higher than room temperature, making it a potential application in magnetoresistive [5] and spintronics [6] devices.

Besides experimental works [3,4,7], various first-principles calculations [3,4,8–13] have been performed to investigate the structural, electronic and magnetic properties of the bulk Sr_2FeMoO_6. It has been found that, the crystalloid structure of the Sr_2FeMoO_6 is a body-centered tetragonal (BCT) with a space group of I4/mmm (No. 139). The single perovskite units $SrFeO_3$ and $SrMoO_3$ alternate along three crystallographical axes. The corners of each single perovskite unit are in turn occupied by the Fe and Mo with oxygen atoms located in between, forming alternate FeO_6 and MoO_6 octahedra along the three cubic axes. The Sr atoms occupy the hollow formed by the corners of FeO_6 and MoO_6 octahedra at the body-centered positions. The Fe^{3+} ($3d^5$) ion is lowly ionized at a high-spin state of S = 5/2, while the Mo^{5+} ($4d^1$) ion is highly ionized with a low-spin state of S = 1/2. Each of the two TM sublattices is in an FM arrangement, while the two sublattices are in an antiferromagnetic (AFM) coupling, giving rise to the total magnetic moment of 4 μ_B per formula unit (f.u.). The smaller saturation magnetization of 3 μ_B/f.u. at 4.2 K [4], is attributed to the mis-site-type disorder on the TM sites [14–16].

Because the physical and chemical properties of the double perovskites $A_2BB'O_6$ are not only depended on the TM elements B and B′ but also depended on the A element [17,18]. Considering the comparable crystal radius of Pb^{2+} (1.63 Å) with that of Sr^{2+} (1.58 Å) each ion with a 12-fold oxygen coordination [19], in our previous paper [20], we proposed to substitute Sr^{2+} ion with Pb^{2+} ion in Sr_2FeMoO_6 and the structural, electronic and magnetic properties of the bulk Pb_2FeMoO_6 have been studied in detail. The HM-FM nature implies a potential application of this new compound in magnetoresistive and spintronics devices. One year later, the magnetic properties and magnetoresistive effects of the Pb_2FeMoO_6 were systematically studied in experiment [21]. Recently, the HM-FM behavior has also been observed in numerous double perovskites, such as Sr_2CoFeO_6 [22], Ba_2CrMoO_6 and Ba_2FeMoO_6 [23], and which also present completely spin polarization of the conduction electrons crossing the Fermi level and thus offer potential technological applications in the devices of single-spin electron source and high-efficiency magnetic sensor.

However, these devices are usually constructed by two HM-FM films (e.g. Pb_2FeMoO_6) sandwiching a thin nonmagnetic film (e.g. Cu film in

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spin valve device) or a very thin insulating layer (e.g. Al_2O_3 film in magnetic tunnel junction device) [6]. So maintaining the HM-FM nature in Pb_2FeMoO_6 film is a key issue for its technological applications in magnetoresistive and spintronics devices and just the aim of this work to check the electronic and magnetic properties of a thin Pb_2FeMoO_6 film. It is found that the three possible terminations of the (001)-oriented thin films of the double perovskite Pb_2FeMoO_6 maintain the HM-FM character and thus can be potentially utilized and applied to the technological applications in magnetoresistive and spintronics devices. The rest of the paper is organized as follows. In Section 2, the calculation methods and models are described in detail. The results and discussions are given in Section 3 and the last section is devoted to the conclusions.

2. Calculation methods and models

The calculations are performed using the Vienna ab-initio simulation package (VASP) based on the density function theory (DFT) [24–27]. The interactions between electron and ionic core are represented by the projector augmented wave (PAW) potentials [28]. The exchange and correlation of the electrons are treated by the Perdew-Burke-Ernzerhof (PBE) [29] formulation of the generalized gradient approximation (GGA) taking into account on-site Coulomb repulsive energy U (GGA + U) (U = 2.0 eV for Fe and 1.0 eV for Mo) [9,30], since both Fe and Mo atoms have localized 3*d* and 4*d* electrons, respectively. To relax the ions into their ground states and thus to obtain the optimized structures, a conjugate-gradient algorithm is used and the energies and the forces on each ion are converged within 1.0×10^{-4} eV/atom and 0.02 eV/Å, respectively. The cutoff energy of the plane-wave set is chosen to be 450 eV. The Pb $6s^2 6p^2$, Fe $3d^6 4s^2$, Mo $4d^5 5s^1$ and O $2s^2 2p^4$ electrons are treated as valence electrons. The k-point is sampled according to the Monkhorst-Pack scheme [31] with special *k* points of $11 \times 11 \times 1$, together with a Gaussian smearing broadening of 0.1 eV.

From optimized structure of the bulk Pb_2FeMoO_6 [20], we know that the interlayer spacing of c/4 = 7.94/4 = 1.985 Å is larger than that of a/4 = b/4 = 5.60/4 = 1.40 Å and the (001)-oriented thin film consists of alternately neutral $FeMoO_4$ and PbO layers along c-axis, as is shown in the left panels of Fig. 1, three possible terminations of the (001)-oriented thin film of Pb_2FeMoO_6 of (a) 10-layer asymmetric but stoichiometric (five formula units both $FeMoO_4$ and PbO terminated) (Pb:10, Fe:5, Mo:5, O:30) (shortly, 10-L $FeMoO_4$ and PbO terminated), (b) 9-layer symmetric but nonstoichiometric $FeMoO_4$ terminated (Pb:8, Fe:5, Mo:5, O:28) (9-L $FeMoO_4$ terminated), and (c) 11-layer symmetric but nonstoichiometric PbO terminated (Pb:12, Fe:5, Mo:5, O:32) (11-L PbO terminated) are considered here for completeness. Each thin film is separated by a 10 Å vacuum region along c-axis to avoid interaction between thin film and its periodic images.

3. Results and discussions

3.1. Optimized structures

The optimized structures of the 10-L $FeMoO_4$ and PbO terminated, 9-L $FeMoO_4$ terminated and 11-L PbO terminated thin films of the Pb_2FeMoO_6 are shown in left panels of Fig. 1(a), (b) and (c), respectively. The corresponding bond lengths of the Fe—O and Mo—O bonds along Fe—O—Mo—O—Fe chain or Mo—O—Fe—O—Mo chain parallel to and perpendicular to the film surface are shown in right panels of Fig. 1(a), (b) and (c) within (110) plane. The detail fractional coordinates *x*, *y* and *z* along a, b and c axes for inequivalent Pb, Fe, Mo and O atoms on inequivalent layers are summarized in Tables 1, 2 and 3 together with partial magnetic moments μ_s, μ_p and μ_d as well as total

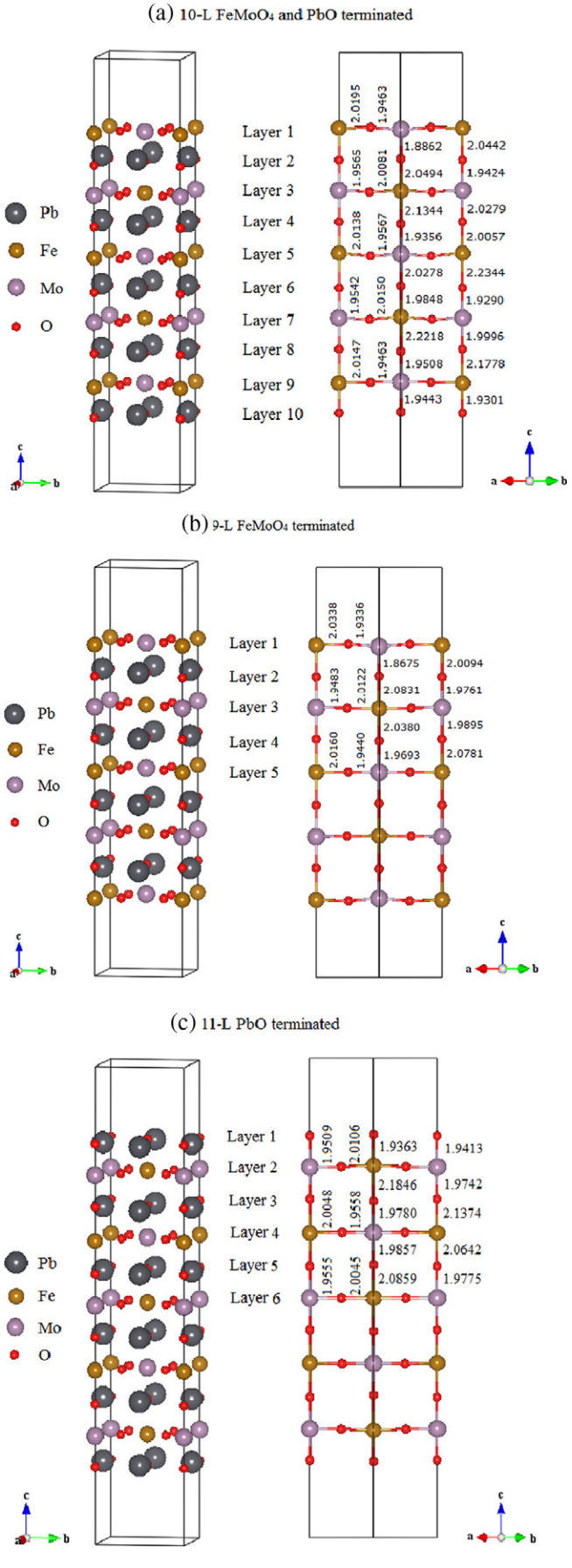

Fig. 1. The optimized structures (left panels) and the Fe—O and Mo—O bond lengths (in Å) along Fe—O—Mo—O—Fe chain or Mo—O—Fe—O—Mo chain within (110) plane (right panels) of (a) 10-L $FeMoO_4$ and PbO terminated, (b) 9-L $FeMoO_4$ terminated and (c) 11-L PbO terminated (001)-oriented thin films of double perovskite Pb_2FeMoO_6.

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