様　式　C－１９、F－１９－１、Z－１９　（共通）

科研費
KAKENHI

科学研究費助成事業　　研究成果報告書

平成 ３０ 年　 ５ 月　 １ 日現在

機関番号：１１３０１
研究種目：研究活動スタート支援
研究期間：2017～2017
課題番号：１７Ｈ０６５１３

研究課題名（和文）Thin films and devices based on fully compensated half-metallic ferrimagnetic materials

研究課題名（英文）Thin films and devices based on fully compensated half-metallic ferrimagnetic materials

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交付決定額（研究期間全体）：（直接経費）　　1,100,000円

研究成果の概要（和文）：本研究は、磁化補償されたフェリ磁性体ハーフメタルホイスラー合金Mn1.5FeV0.5Alの高規則度単結晶薄膜の成長を目的とし、最終的にはその強磁性トンネル接合への応用を目指すものである。スパッタ法によりL21規則構造を有するMn1.5FeV0.5Alの単結晶薄膜の作製を達成した。その材料薄膜がネールのP型の温度依存性を示すフェリ磁性を有することが異常ホール効果測定から明らかとなった。また、異常磁気抵抗効果は負の値を示しハーフメタル性を示唆した。これらの成果は、この材料薄膜が将来のスピントロニクス応用へ向けた磁気トンネル接合のための有望な電極材料であることを実証するものである。

研究成果の概要（英文）：One goal in this research project was to grow and extensively investigate the thin films of the new class of material: fully compensated half-metallic ferrimagnets Mn1.5FeV0.5Al. Final goal is to create novel magnetic tunnel junctions with this new material for advanced spintronic applications. We succeeded, for the first time, to grow epitaxial single-crystalline Mn1.5FeV0.5Al thin films with highly L21-ordered Heusler structure prepared using an ultrahigh vacuum magnetron sputtering system. The film exhibited a Neel P-type ferrimagnetic character in AHE, where the magnetic moment of the antiparallel aligned sublattices is always larger in one direction than in the opposite one and no compensation points were obtained. A negative anisotropic magnetoresistance (AMR) ratio was observed. This negative sign of AMR proved the half-metallicity of the films. The result demonstrated that the material films were very promising for advanced spintrnoic applications.

研究分野：Spintronics

キーワード：Spintronics　Heusler alloys　ferrimagnets

１．研究開始当初の背景

Recently, zero net magnetic moment materials, such antiferromagnet (AFM), have attracted much interest as future perspective materials for spintronics [1]. The fully compensated half-metallic ferrimagnet (FCHMF) is a new class of material combining zero net magnetic moments and fully spin-polarized band structure [2,3], which can drastically improve the present spintronics devices.

Very recently, we realized the FCHMF properties only in the bulk polycrystalline Heusler compound $Mn_{1.5}FeV_{0.5}Al$ [2], however not in thin films or devices, so far.

２．研究の目的

The goal in this research project was to growth and investigates extensively the new class of material: fully compensated half-metallic ferrimagnets (FCHMF) $Mn_{1.5}FeV_{0.5}Al$ as thin films. The final goal is to create novel magnetic tunnel junction with this new materials for advanced spintronic applications.

$Mn_{1.5}FeV_{0.5}Al$ crystallizes in the regular $L2_1$ cubic Heusler structure with space group 225. The Al atoms occupy the 4*a* position; the *4b* positions are equally occupied by V and Mn atoms and a statistical distribution of Mn and Fe atoms at *8c* (Fig. 1c).

The magnetic moments on the different sublattice couple antiparallel. The Mn moment on *4b* has a localized moment and is compensated by the magnetic moments of the remaining *8c* sublattice moments (Fe, Mn).

３．研究の方法

All the samples were prepared using an ultrahigh vacuum (UHV) magnetron sputtering system with a base pressure of less than 3×10^{-7} Pa, located in Mizukami Lab. at WPI Advanced Institute for Materials Research (WPI-AIMR) in Tohoku University.

We have grown $Mn_{1.5}FeV_{0.5}Al$ thin films directly on single crystalline MgO(001) substrates, because the lattice mismatch between $Mn_{1.5}FeV_{0.5}Al$ and MgO is 2.2%. For the deposition, a off-stoichiometric quaternary target of $Mn_{37.5}Fe_{25}V_{12.5}Al_{25}$ was used, and the films were capped by 2-nm-thick Ta or Al to prevent the oxidation of the films. The composition of the deposited films was $Mn_{36.1}Fe_{27.5}V_{11.9}Al_{24.5}$, which was measured by inductively coupled plasma mass spectrometry (ICP-MS).

The crystal structures and lattice parameters of the samples were determined using an X-ray diffractometer (XRD) with Cu K_α radiation. Micron-sized bar Hall bars (120×24 μm^2) for the Hall and longitudinal resistance measurements were fabricated by the conventional ultraviolet photolithography and Ar ion milling.

The electrical transport properties were investigated with a physical property measurement system (PPMS, Quantum Design) using a four-probe method in a temperature range of 5 - 400 K under an applied magnetic field up to 90 kOe.

The Hard X-ray photoelectron spectroscopy (HAXPES) experiment was performed at the beamline BL47XU of SPring-8 using a photon energy of 7.9 keV for excitation and a temperature of 14 K.

４．研究成果

We succeed, for the first time, to grow the epitaxial single-crystalline fully compensated half-metallic ferrimagnet $Mn_{1.5}FeV_{0.5}Al$ thin films with highly $L2_1$-ordered Heusler structure and improved their half -metallicity.

The 30-nm-thick thin films were deposited at various substrate temperatures T_s starting from room temperature (RT) to 700°C. XRD patterns of the films prepared with different T_s are shown in Fig. 1. With increasing the substrate temperature T_s, the intensity of diffraction peaks (004) is enhanced, and the (002) superlattice diffraction peak from the *B*2 structure appears for films deposited at $T_s \geq 500$°C.

The (111) superlattice diffraction peak from the $L2_1$ structure ordering was clearly observed from the ϕ-pole figures, as shown in Fig. 1 (b). The (111) peaks with four-fold symmetry are observed with shifts of 45° relative to the MgO (111) peaks and proves the epitaxial growth of $Mn_{1.5}FeV_{0.5}Al$ with the relationship: $Mn_{1.5}FeV_{0.5}Al$ (001)[100]//MgO(001)[110]. The lattice parameter *a* and *c* for the film grown at 600°C are determined to $a = 5.853$ Å and $c = 5.822$ Å, respectively.

Bulk sensitive hard X-ray photoelectron spectroscopy of the core levels in $Mn_{1.5}FeV_{0.5}Al$ was measured to investigate the chemical ordering and the electronic properties of the films. The spectra of the Fe *2p*, V *2p*, and Mn *2p* core levels are shown in Fig. 2a to 2c, respectively. All *2p* core states exhibit a clear and sharp spin-orbit splitting into $2p_{1/2}$ and $2p_{3/2}$ sublevels. The shape of the spectra is typical for metallic states and proves that the films are not oxidized.

An additional exchange splitting is observed in of *Mn* $2p_{3/2}$ (inset in Fig. 2c), this is caused by the Coulomb interaction of the *2p* core holes and the localized Mn *3d* valence electrons. Such splitting is not observed in *Fe 2p* and *V 2p* states; this is due to the delocalized *3d* states in Fe and V. The magnitude of splitting in 3s core level peak in transition metal has a direct relation to the spin magnetic moment of the atom. As shown in Fig. 2d, both *Fe3s* and *Mn3s* states exhibit an exchange splitting of about 4 eV. Though, the nearly vanishing net magnetization in $Mn_{1.5}FeV_{0.5}Al$, the magnetic moments of the sublattices is present. This was proven previously in $Mn_{1.5}FeV_{0.5}Al$ bulk by X-ray magnetic circular dichroism measurements [2].

Figure 3a shows the temperature dependence of the anomalous Hall resistivity $\rho^{AHE}(T)$ for the film deposited at T_s = 600°C measured at 16 mT. $\rho^{AHE}(T)$ increases below Curie temperature (T_c = 370 K) reaching a maximum at 250 K. The film exhibit a Néel P-type ferrimagnetic character, where the magnetic moment of the antiparallel aligned sublattices is always larger in one direction than in the opposite one and no compensation point is obtained. The low value of $\rho^{AHE}(T)$ about 400 nΩ · cm, as shown in Fig. 3b, arises due to a low magnetization.

To evaluate the half-metallicity of $Mn_{1.5}FeV_{0.5}Al$, we have measured the anisotropic magnetoresistance (AMR), where the AMR ratio was calculated with $(R(\phi) - R_\perp / R_\perp] \times 100$; ϕ is the angle between the magnetic field and the current, and $R_\perp$ is the resistivity when the magnetization direction is perpendicular to the electric current direction.

As shown in Fig. 4, the AMR exhibits the twofold and fourfold symmetric term, which originated from the intrinsic electronic property and the lattice symmetry. Beside, a negative AMR ratio of - 0.11% was observed at 5 K and a magnetic field of 4 T. The negative sign of AMR proves the half-metallicity of $Mn_{1.5}FeV_{0.5}Al$ film, as discusses in the other half-metallic Heusler alloys, because the density of states in down-spin electrons at the Fermi edge is absent and the resistivity is dominant by the *s-d* scattering of up-spin channel.

Therefore, these results demonstrate that the $Mn_{1.5}FeV_{0.5}Al$ film is a promising electrode film of magnetic tunnel junctions for advanced spintronic applications.

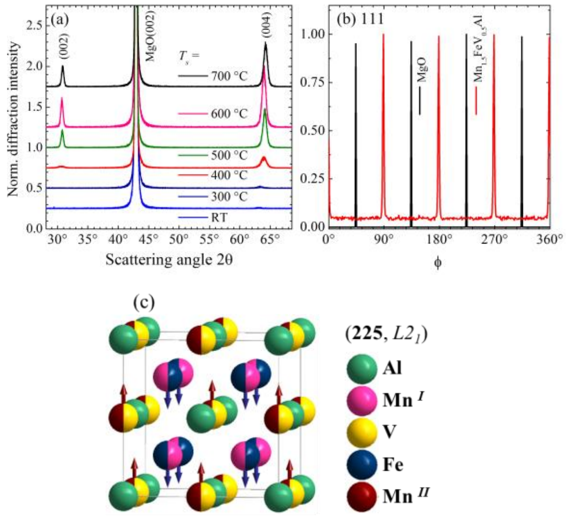

Figure 1: (a) θ-2θ X-ray diffraction spectra of 30nm-Heusler films $Mn_{1.5}FeV_{0.5}Al$ deposited at different substrate temperatures T_s , and (b) ϕ-scan measurements of the (111) planes from the MgO substrate and $Mn_{1.5}FeV_{0.5}Al$ film deposited at 600°C. (c) The $L2_1$ unit cell of $Mn_{1.5}FeV_{0.5}Al$ Heusler structure.

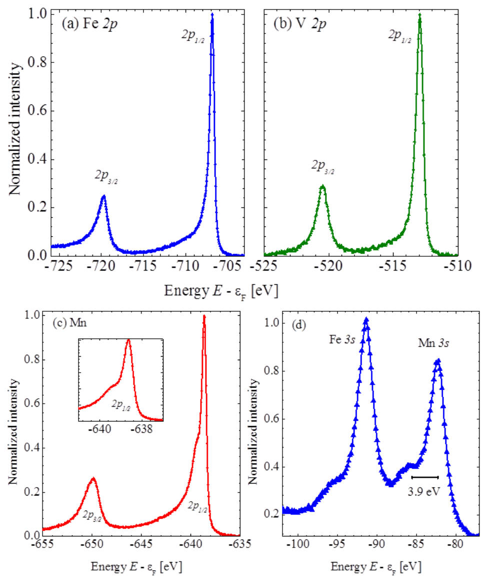

Figure 2: High-energy photoelectron spectra of the (a) *Fe 2p*, (b) *V 2p*, (c) *Mn 2p*, and (d) *Fe 3s, Mn3s* core level of $Mn_{1.5}FeV_{0.5}Al$ thin film. The inset in (c) shows the zoom of the *Mn* $2p_{3/2}$ state.

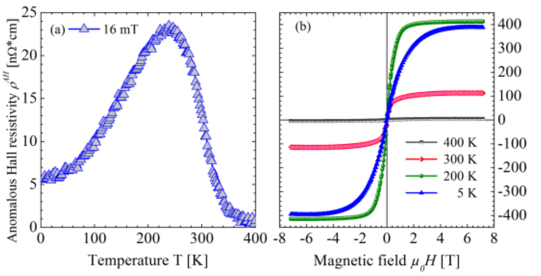

Figure 3: Temperature dependence of Hall resistivity measured at 16 mT (a), and field dependence of the Hall effect measured at different temperatures (b).

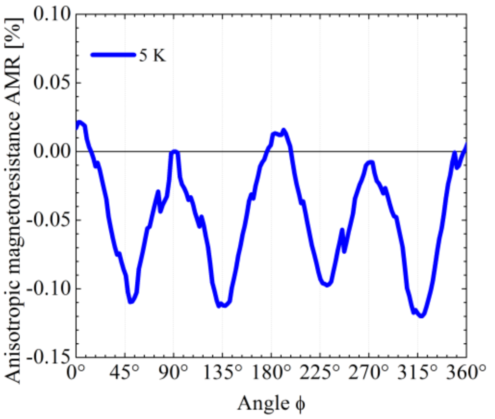

Figure 4: Dependence of the AMR ratio on the in-plane relative angle ϕ in $Mn_{1.5}FeV_{0.5}Al$ film measured at 5 K.

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[1] S. Fukami, C. Zhang, S. DuttaGupta, A. Kurenkov, and H. Ohno, *Nature Mater.* **15**, 535 (2016).
[2] R. Stinshoff, A. K. Nayak, G. H. Fecher, B. Balke, S. Ouardi et al., *Phys. Rev. B* **95** 060410(R) (2017).
[3] M. E. Jamer et al., *Phys. Rev. Appl.* **7**, 064036 (2017).

５．主な発表論文等
（研究代表者、研究分担者及び連携研究者には下線）

〔雑誌論文〕（計　０件）

〔学会発表〕（計　２件）
[1] “Epitaxial growth of compensated ferrimagnetic Heusler thin films Mn-Fe-V-Al (Oral)”, Siham Ouardi, Kazuya Z. Suzuki, and Shigemi Mizukami, Joint spring meeting of German Physical Society DPG and the European Physical Society EPS Condensed Matter Divisions, Berlin, Germany, Mar. 11th - 16th, 2018.
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〔図書〕（計　０件）

〔産業財産権〕

○出願状況（計　０件）

名称：
発明者：
権利者：
種類：
番号：
出願年月日：
国内外の別：

○取得状況（計　０件）

名称：
発明者：
権利者：
種類：
番号：
取得年月日：
国内外の別：

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