様　式　Ｃ－１９、Ｆ－１９－１、Ｚ－１９　（共通）

科学研究費助成事業　　研究成果報告書

令和　５ 年　 ５ 月 ２３ 日現在

機関番号：８２５０２
研究種目：研究活動スタート支援
研究期間：2021～2022
課題番号：２１Ｋ２０５０８

研究課題名（和文）Exploring novel graphene/ferrimagnetic Heusler alloy heterostructures for spin-photonic applications

研究課題名（英文）Exploring novel graphene/ferrimagnetic Heusler alloy heterostructures for spin-photonic applications

研究代表者
ＢＥＮＴＬＥＹ　ＰＨＩＬＬＩＰ．ＤＡＶＩＤ（BENTLEY, PHILLIP DAVID）

国立研究開発法人量子科学技術研究開発機構・高崎量子応用研究所　量子機能創製研究センター・博士研究員

研究者番号：４０９０６２６０

交付決定額（研究期間全体）：（直接経費）　　2,100,000円

研究成果の概要（和文）：スピンフォトニクスへの応用を視野に二次元物質/フェリ磁性ホイスラー合金ヘテロ構造の研究に取り組んだ。その中で、新規立方晶フェリ磁性ホイスラー合金Mn2FeGa薄膜の開発に成功した。Mn2FeGa薄膜は、強い一軸磁気異方性による巨大な歪誘起垂直磁気異方性や高い化学規則度による高スピン偏極率など、MRAMをはじめスピントロニクス、スピンフォトニクスデバイスへの応用に有望な物性を持つことを明らかにした。これらの成果は、学術誌(Physical Review Materials)への論文投稿(査読中、受理見込み)や学会講演、特許申請を行った。

研究成果の学術的意義や社会的意義
Cubic MFG extraordinary magnetic properties such as its large PMA and high spin polarization make it a very suitable candidate for spintronic application such as MRAM. This will help us overcome current limits on magnetic memory and MFG is likely to be a leading material in next-generation memory.

研究成果の概要（英文）：I explored and developed a novel cubic phase of the ferrimagnetic Heusler compound Mn2FeGa (MFG) with the ultimate aim of coupling this material with 2D materials such as graphene for spin photonic applications. This newly developed material shows several favourable properties for spintronic applications including: significant perpendicular magnetic anisotropy (PMA), high chemical ordering, and high spin polarization. These properties demonstrate a material suitable for spintronic applications such as magnetic random-access memory (MRAM). These results have led to a publication which is currently under review, receiving positive feedback and is soon to be published. Furthermore, these results have led to a patent for cubic MFG/Cr/MgO buffer layers which is currently in press. Finally, these results have been presented at several national conferences and have attracted significant attention among the spintronics community.

研究分野：spintronics

キーワード：Heusler Compound　Ferrimagnetism　PMA　Half-metallicity　MRAM

１．研究開始当初の背景

With the emergence of the Internet of Things (IoT), there has been a drastic increase in the demand for increased speed and capacity of information transferred by networks. To compete with this demand, different forms of memory are required such as magnetic random-access memory (**MRAM)**, a non-volatile, low-energy, and high-speed sources of memory will help in overcoming the challenges faced with current volatile electronic sources of memory. However, MRAM **requires** magnetic materials which show high spin polarization (SP) and large perpendicular magnetic anisotropy (PMA).

Over the past couple of decades Heusler alloys have attracted increasing attention for their use in MRAM due to observations of high spin-polarization (Y. Sakuraba *et al*., Appl. Phys. Lett., **88**, (2006)) and low damping constants (C. Guillemard *et al*., Appl. Phys. Lett., **88**, (2006)). However, whilst ferromagnetic Heusler alloys such as $Co_2FeGa_{0.5}Ge_{0.5}$ have been shown to be highly spin-polarized (K. Hono *et al*., Acta Mater., **60**, (2012)), Heusler alloys also often exhibit an in-plane magnetization which significantly affects their performance in MRAM. Ferrimagnetic cubic Heusler alloys in comparison to their ferromagnetic counterparts have shown weak PMA as well high spin polarization (H. Kurt *et al*., Phys. Rev. Lett., **112**, (2014)). If high spin-polarization and large PMA can be achieved in a cubic ferrimagnetic Heusler alloy, such a material would be suitable candidate for applications such as MRAM.

２．研究の目的

The objectives of this project were to: (1) prepare high-quality thin films of the ferrimagnetic Heusler alloy Mn_2Fe_xGa (MFG) in the novel cubic phase, (2) analyse their electronic and magnetic properties using a variety of both experimental and theoretical techniques, and (3) optimise the magnetic properties of these thin films for their use in MRAM.

３．研究の方法

Thin films of MFG were prepared in an ultra-high vacuum (UHV) via magnetron co-sputtering with the composition being controlled by varying the power applied to either the Mn_2Ga or Fe target. These MFG thin films were grown both directly on-top of MgO(001) substrates or a Cr buffer layer, with the latter being the preferred method of growth. The properties of these thin films were analysed using a variety of techniques including X-ray diffraction (XRD), SQUID magnetometry, transmission electron microscopy (TEM) as well as synchrotron radiation-based techniques such as synchrotron Mössbauer spectroscopy performed at BL11XU beamline of the SPring-8 research facility. The electronic structure of MFG was calculated by Keisuke Masuda at the National Institute for Materials Science using density-functional theory (DFT) and second-order perturbation analysis.

４．研究成果

(1) The structure and quality of the MFG thin films were analysed using reflection high-energy electron diffraction (RHEED), TEM, and XRD. As seen via bright-field cross-sectional TEM, epitaxial single-crystalline growth with no sign of interfacial mixing was observed in cubic MFG thin films grown on a Cr buffer layer. The interface between each layer is found to be atomically sharp. Using XRD it was found that, depending on the Fe concentration, either a mixed phase ($x < 1.0$) or a newly developed single cubic phase ($x \geq 1.0$) was observed. In these cubic thin films, a significant tetragonal distortion of $c/a = 1.04 – 1.06$ was induced by the lattice matching between cubic MFG and Cr under

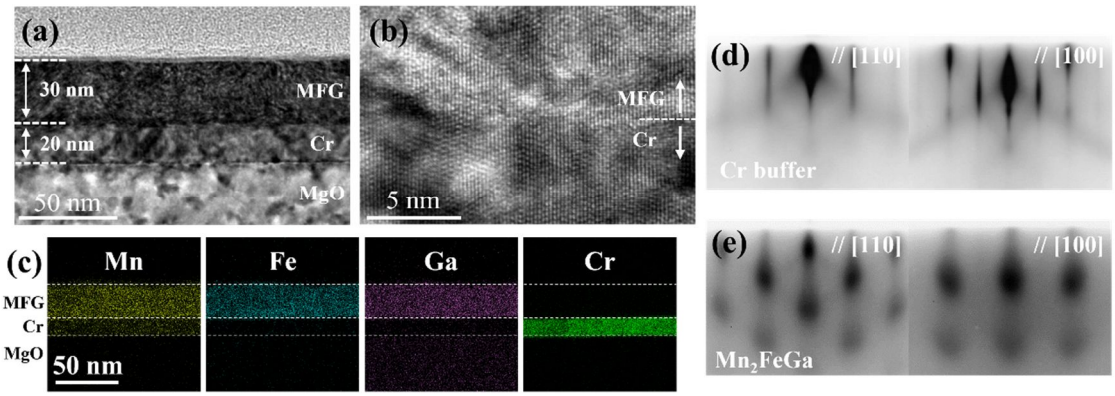

図１ **(a)** Bright-field cross-sectional TEM for the stoichiometric MFG/Cr buffer layer sample, **(b)** high-resolution image, **(c)** EDS mapping of the image shown in **(a)**, **(d)** and **(e)** RHEED of the Cr buffer layer and the stoichiometric MFG thin film, respectively.

the epitaxial relationship. However, the cubic nature of this alloy is preserved under the tetragonal strain as indicated by the presence of other XRD reflections such as the (111) reflection. ϕ-scans of the samples further confirm the epitaxial growth of cubic MFG. For thin films grown directly on top of MgO(001) no tetragonal strain was observed and the quality of growth was found to be much poorer than when grown on a Cr buffer layer.

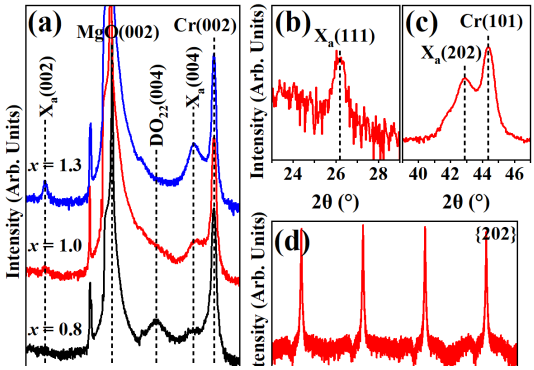

図 2 **(a)** XRD profiles of MFG/Cr buffer samples with different Fe concentrations *x*. XRD of the stoichiometric MFG/Cr sample

(2) The magnetic properties of these thin films were evaluated using SQUID magnetometry and synchrotron Mössbauer spectroscopy. In the stochiometric MFG/Cr sample, SQUID revealed that these thin films exhibited PMA which exceeded 0.75 MJ/m^3 which is a result of the significant tetragonal strain induced by the Cr buffer layer. This PMA is the largest value reported to date for any cubic ferrimagnetic Heusler alloy, far greater than that observed Mn_2RuGa or Mn_2CoGa (P. D. Bentley *et al*., Phys. Rev. Mater., under review). In the stoichiometric cubic MFG/Cr, SQUID results also show a material with a low saturation magnetization ideal for fast switching in MRAM as well as good squareness of the magnetization curve. Synchrotron Mössbauer spectroscopy showed the internal hyperfine magnetic field of Fe to be small in cubic MFG with a magnetic moment of ~0.25 μ_B being observed in the stoichiometric MFG/Cr sample. This result closely matches the theoretically predicted value of 0.22 μ_B estimated for the X_a cubic inverse full-Heusler crystal structure of MFG. Therefore, these results suggest that cubic MFG grown on a Cr buffer layer possesses a high degree of chemical ordering close X_a crystal structure and therefore is likely to be highly spin polarized.

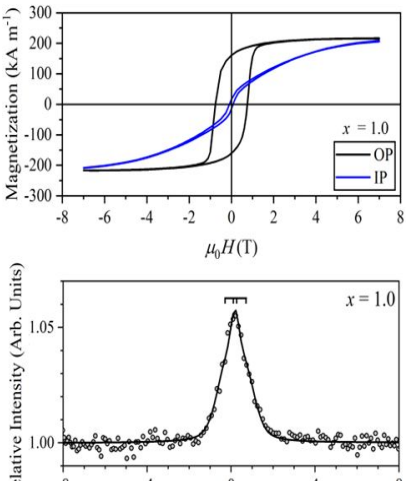

図 3 (upper panel) In-plane (IP) and out-of-plane (OP) magnetic hysteresis loops measured at room temperature for cubic MFG/Cr/MgO. (lower panel) Mössbauer spectrum of the same sample.

(3) The density of states (DOS) of the unstrained and strained case of cubic MFG were calculated showing that even with a significant tetragonal strain of 4.1% (as measured for the stoichiometric MFG/Cr sample) that the half-metallic nature of MFG is preserved. First-principles calculations revealed that the large PMA observed in cubic MFG originates from specific features of Fe *d* orbitals around the Fermi level, which give rise to a large uniaxial magnetocrystalline anisotropy under the strain. This is a unique characteristic of cubic MFG as no other Y element in a Mn_2YZ-type Heusler alloy possesses such similar electronic features around the Fermi level.

(4) Thanks to the funding of this project, a new cubic crystal phase of MFG was successfully developed. Cubic MFG with its small saturation magnetization, large PMA, and possibility of being highly spin-polarized make it an ideal candidate for the development of MRAM and other spintronic devices.

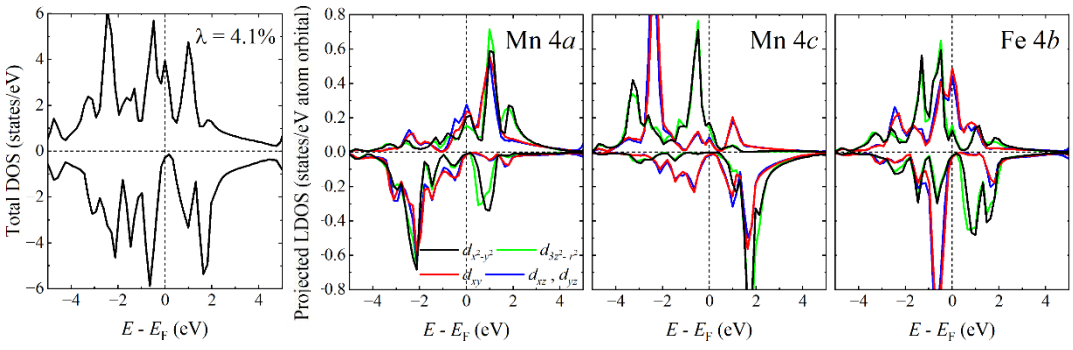

図 4 Total DOS and projected local DOS of X_a-ordered MFG with a tetragonal strain of 4.1%

５．主な発表論文等

〔雑誌論文〕　計1件（うち査読付論文　1件／うち国際共著　1件／うちオープンアクセス　1件）

１．著者名 P. D. Bentley, S. Li, K. Masuda, Y. Miura, Y. Du, T. Mitsui, K. Fujiwara, Y. Kobayashi, T. Y. Guo, G. Q. Yu, C. Suzuki, S. Yamamoto, F. Zheng, Y. Sakuraba, and S. Sakai	４．巻 -
２．論文標題 Cubic-type Heusler compound Mn2FeGa thin film with strain-induced large perpendicular magnetic anisotropy	５．発行年 2023年
３．雑誌名 Physical Review Materials	６．最初と最後の頁 -
掲載論文のDOI（デジタルオブジェクト識別子） なし	査読の有無 有
オープンアクセス オープンアクセスとしている（また、その予定である）	国際共著 該当する

〔学会発表〕　計2件（うち招待講演　0件／うち国際学会　0件）

１．発表者名 Phillip David Bentley, Chihiro Suzuki, Shunya Yamamoto, Songtian Li, and Seiji Sakai
２．発表標題 Cubic Ferrimagnetic Full-Heusler Mn2FexGa Thin Films with Strong Perpendicular Magnetic Anisotropy
３．学会等名 The 69th JSAP Spring Meeting 2022
４．発表年 2022年

１．発表者名 Phillip David Bentley, Songtian Li, Tengyu Guo, Guoqiang Yu, Takaya Mitsui, Yasuhiro Kobayashi, Kosuke Fujiwara, and Seiji Sakai
２．発表標題 Revealing the Local Magnetic Properties of Cubic Ferrimagnetic Full-Heusler Alloys Using Synchrotron Mossbauer Spectroscopy
３．学会等名 The 83rd JSAP Autumn Meeting 2022
４．発表年 2022年

〔図書〕　計0件

〔出願〕　計0件

〔取得〕　計1件

産業財産権の名称 鉄含有擬立方晶マンガン基フェリ磁性ホイスラー合金、及び、これを用い た磁気抵抗記憶素子	発明者 境、ベントレー、李、三井、藤原、増田、三浦	権利者 量子科学技術研究開発機構
産業財産権の種類、番号 特許、特願2023-027745	取得年 2023年	国内・外国の別 国内

〔その他〕

―

６．研究組織

	氏名 （ローマ字氏名） （研究者番号）	所属研究機関・部局・職 （機関番号）	備考

７．科研費を使用して開催した国際研究集会

〔国際研究集会〕　計0件

８．本研究に関連して実施した国際共同研究の実施状況

共同研究相手国	相手方研究機関