HOKKAIDO UNIVERSITY

Title	Effect of off-stoichiometry on the half-metallic character of quaternary Heusler alloy Co2(Mn,Fe)Si [an abstract of dissertation and a summary of dissertation review]
Author(s)	Ayele, Kidist Moges
Degree Grantor	北海道大学
Degree Name	博士(工学)
Dissertation Number	甲第12401号
Issue Date	2016-09-26
Doc URL	https://hdl.handle.net/2115/63327
Rights(URL)	https://creativecommons.org/licenses/by-nc-sa/2.1/jp/
Type	doctoral thesis
File Information	Ayele_Kidist_Moges_abstract.pdf, 論文内容の要旨

Hokkaido University Collection of Scholarly and Academic Papers : HUSCAP

学 位 論 文 内 容 の 要 旨

博士の専攻分野の名称　博士（工学）　氏名　Ayele Kidist Moges

学 位 論 文 題 名

Effect of off-stoichiometry on the half-metallic character of quaternary Heusler alloy Co_2(Mn,Fe)Si
（4 元ホイスラー合金 Co_2(Mn,Fe)Si のハーフメタル性に対する非化学量論的組成の影響）

The search for electronic devices with better functionalities such as nonvolatility, reconfigurable logic functions, and low power consumption led to a novel area of electronics, called spintronics. Spintronic devices utilize the spin degree of freedom of the electron in addition to its charge. Thus, the creation of a highly spin-polarized current is essential for spintronic devices. Originating from the existence of an energy gap at the Fermi level (E_F) for one spin direction (mostly the minority-spin band), half-metallic ferromagnets (HMFs) feature complete spin polarization at E_F. Accordingly, HMFs are seen as one of the most suitable spin source materials for spintronic devices. Co-based Heusler alloys (Co_2YZ, where $Y$is usually a transition metal and Z is a main group element) are among the most extensively applied to spintronic devices, including magnetic tunnel junctions (MTJs), in particular those with a MgO tunnel barrier, giant magnetoresistance (GMR) device, and to spin injection into semiconductors. One of the notable advantages of Co_2YZ is that they feature a variety of materials, including not only ternary alloys but also quaternary alloys, thereby providing flexibility in their application to spintronic devices.

One quaternary Heusler alloy system, $Co_2(Mn_{1-x}Fe_x)Si$, has been studied from the view point of tuning the E_F position around the center of the half-metal gap by changing the composition x in $Co_2(Mn_{1-x}Fe_x)Si$ in order to control the valence electron number per formula unit, Z_t. In practice, off-stoichiometry in Co_2YZ thin films prepared by magnetron sputtering or molecular beam epitaxy is inevitable and leads to structural defects. If the minority-spin in-gap states are introduced at E_F by a structural defect for a half-metallic thin film, the half-metallicity is strongly degraded. Thus, it is important to understand the effect of defects associated with off-stoichiometry on half-metallicity in order to take full advantage of the half-metallic character of Co-based Heusler alloys.

H. Liu et al. from our research group recently investigated the influence of varying the film composition of Co_2(Mn,Fe)Si (CMFS) Heusler alloy on the tunneling magnetoresistance (TMR) characteristics of fully epitaxial CMFS/MgO/CMFS MTJs (CMFS MTJs) with $Co_2(Mn_{\alpha'}Fe_{\beta'})Si_{0.84}$ electrodes. Through this study, we demonstrated giant TMR ratios of 2610% at 4.2 K and 429% at 290 K for CMFS MTJs with Mn-rich, lightly Fe-doped CMFS electrodes.

The main purpose of the present study was to clarify the effect of off-stoichiometry on the half-metallicity of quaternary Heusler alloy CMFS. To do this, we experimentally investigated the film composition dependence of the saturation magnetization per formula unit, μ_s, of CMFS films with various compositions of α' and β' in $Co_2(Mn_{\alpha'}Fe_{\beta'})Si_{0.84}$, and that of the TMR ratio of CMFS MTJs, in combination with first-principles calculations. Furthermore, in order to clarify the origin of the giant TMR ratios demonstrated for CMFS MTJs with Mn-rich, lightly Fe-doped CMFS electrodes, we experimentally investigated how the TMR ratio of CMFS MTJs with lightly Fe-doped CMFS electrodes depends on the Mn composition α' in $Co_2(Mn_{\alpha'}Fe_{0.16})Si_{0.84}$ electrodes in the Mn-rich composition region. Finally, in this dissertation, we investigated the bias voltage dependence of the spin-dependent differential tunneling conductance of the CMFS and CMS MTJs along with its origin. To do this, we

studied the dI/dV versus V characteristics (= G spectra) of MTJs having CMFS and CMS electrodes that showed giant TMR ratios.

This dissertation is organized into six chapters and the main content of each chapter is as follows:

Chapter 1 gives the general introduction and background of this dissertation. It first reviews important experimental and theoretical backgrounds related to this study, details the experimental investigation and result of the influence of film composition on the TMR characteristics of fully epitaxial CMFS MTJs and then it provides the overall purpose and approaches of this dissertation.

Chapter 2 describes the overall experimental and computational methods followed in this dissertation. It first details the sample preparation, structural characterization and measurement techniques of the samples prepared in this study and then it explains the theoretical and computational methods.

Chapter 3 details the effect of off-stoichiometry on the half-metallic character of CMFS investigated by studying the film composition dependence of μ_s and the TMR ratio for two series of CMFS films. The experimentally investigated μ_s and TMR ratios in combination with first-principles calculations indicated that $Co_{Mn/Fe}$ antisites in the quaternary alloy are detrimental to half-metallicity, in a similar way as in ternary alloy CMS. It was demonstrated that (Mn+Fe)-rich compositions are highly effective at suppressing harmful $Co_{Mn/Fe}$ antisites and that a relatively small Fe ratio in the total (Mn+Fe) composition retains a half-metallic state.

Chapter 4 clarifies the origin of the giant TMR ratio obtained for CMFS MTJs with Mn-rich, lightly Fe-doped CMFS electrodes through the dependence of the TMR ratio of CMFS MTJs with $Co_2(Mn_{\alpha'}Fe_{0.16})Si_{0.84}$ electrodes on α' in the Mn-rich composition region. It was revealed that residual $Co_{Mn/Fe}$ antisites were further suppressed in lightly Fe-doped CMFS due to the fact that the available (Mn+Fe) composition in (Mn+Fe)-rich CMFS became larger than the critical Mn composition in Mn-rich CMS. This resulted in giant TMR ratios for CMFS MTJs with Mn-rich, lightly Fe-doped CMFS electrodes. Furthermore, it was revealed from the dI/dV versus V characteristics that the Fermi level position of the Mn-rich, lightly Fe-doped CMFS was pushed up to around the center of the half-metal gap by an increase in the Z_t value due to small Fe doping compared with Mn-rich Co_2MnSi. This result supported our finding of the enhanced half-metallicity of the Mn-rich, lightly Fe-doped CMFS obtained by the film composition dependence of the TMR ratio.

Chapter 5 examines the bias voltage dependence of the spin-dependent differential tunneling conductance of CMFS and CMS MTJs along with the origin of this dependence. Clear dip structures were found in the G spectra for the antiparallel magnetization alignment (AP) in a small bias region of up to ±70 mV. Moreover, it is found that the dI/dV value for AP at characteristic voltages, V_s = ±70 mV, normalized by the value at V = 0 increased linearly with the TMR ratio at 4.2 K. Thus, it was clarified that MTJs having electrodes with higher spin polarization showed stronger bias voltage dependence. This feature was consistently explained by spin-flip tunneling via a magnon excited by a hot electron along with the strong temperature dependence of tunneling conductance for AP by spin-flip tunneling via a thermally excited magnon.

Chapter 6 summarizes and concludes this dissertation.

In summary, the findings of this dissertation indicate that the approach to controlling off-stoichiometry and film composition is promising for fully utilizing the half-metallicity of quaternary CMFS thin films as spin source materials.