

氏名(本籍)	ボラプラガタ ヴアラプラサド (インド)
学位の種類	博士(工学)
学位記番号	博甲第 5652 号
学位授与年月日	平成 23 年 3 月 25 日
学位授与の要件	学位規則第 4 条第 1 項該当
審査研究科	数理物質科学研究科
学位論文題目	<b>Search for highly spin polarized quaternary Heusler alloys</b> (高いスピン分極率をもつ 4 元系ホイスラー合金の探索)

主査	筑波大学教授	Ph. D.	宝野和博
副査	筑波大学教授	工学博士	喜多英治
副査	筑波大学教授	博士(工学)	三谷誠司
副査	筑波大学准教授	博士(工学)	柳原英人

### 論文の内容の要旨

Heusler alloys have attracted great deal of interest because of their half-metallicity with high Curie temperature ( $T_c$ ). Although several ternary Heusler alloys are predicted to have 100% spin polarization, experimentally measured values were much smaller. This thesis reports the results of a thorough materials search for highly spin polarized ferromagnetic Heusler alloys using the point contact Andreev reflection (PCAR) technique. This thesis comprises of 9 chapters. Chapter 1 is an outline and background of the Heusler alloys. In chapter 2, the experimental methods employed in this research are introduced. CoMn-based Heusler alloys are of a particular interest because these alloys have high Curie temperature, which is essential for applications as spin polarized electron sources for practical spintronics devices. The search of CoMn based full Heusler alloys have been reported in chapters 3, 4, 5, and 6. In chapter 3 and 4 we attempted to study the effect of alloying different valence atoms for  $Y = \text{Mn}$  position on structural, magnetic and transport properties of quaternary alloy systems  $\text{Co}_2\text{Mn}_{1-x}\text{Ti}_x\text{Sn}$ , and  $\text{Co}_2\text{Mn}_{1-x}\text{Fe}_x\text{Ga}$  with  $x$  ranging from 0 to 1. In chapter 5 and 6, we focused our study on the effect of the  $Z$  element substitution in  $\text{Co}_2\text{MnGa}_{1-x}\text{Z}_x$  ( $Z = \text{Sn}$  and  $\text{Ge}$ ) on structural, magnetic and transport properties of quaternary alloy,  $x$  ranging from 0 to 1. We also investigated the thin films of the  $\text{Co}_2\text{MnGa}_{0.5}\text{Sn}_{0.5}$  alloy that showed high spin polarization in a bulk alloy. We have succeeded in preparing magnetic tunnel junctions (MTJ) using radical oxidation, to make  $\text{CoFe}/\text{Al-O}/\text{CoFe}$  and  $\text{Co}_2\text{MnGa}_{0.5}\text{Sn}_{0.5}/\text{Al-O}/\text{CoFe}$  junctions. These junctions gave a tunnel magneto resistance (TMR) ratio of 50% and 34% at room temperature respectively. Although expected TMR and GMR ratios are high for  $\text{Co}_2\text{MnGa}_{0.5}\text{Sn}_{0.5}$ , realized values were low because of the oxidation of Heusler alloy during oxidation process in the case of TMR and unable sustain the multilayer structure in case of GMR at higher temperature annealing. In chapter 7, we chose CoFe in contrast with the CoMn system to achieve structural stability in thin film form with high Curie temperatures, and we prepared  $\text{Co}_2\text{FeGe}_{1-x}\text{Si}_x$  alloy series and characterization was done and our results indicated that  $x = 0.75$  has high spin polarization and forms stable  $L2_1$  structure. For this composition we fabricated thin films and measured the structural, magnetic and spin polarization properties. In conclusion we found intermetallic compounds which forms stable  $L2_1$  after melting with

high  $T_c$  around 1000 K like  $\text{Co}_2\text{FeGe}_{0.25}\text{Si}_{0.75}$ ,  $\text{Co}_2\text{MnGa}_{0.25}\text{Ge}_{0.75}$ , and  $\text{Co}_2\text{Mn}_{0.5}\text{Fe}_{0.5}\text{Ga}$  are suitable Ferromagnetic electrodes for device applications. Finally in Chapter 8, we demonstrated some insulating layers as capping layer to reduce surface oxidation of permalloy and  $\text{Co}_2\text{FeAl}_{0.5}\text{Si}_{0.5}$  thin films. Spin polarization measured by PCAR has shown a clear difference in the sample with and without capping layers.

## 審査の結果の要旨

本論文は点接触アンドレーフ反射測定により種々の強磁性 4 元系ホイスラー合金のスピンドル率を測定した結果を報告しており、 $\text{Co}_2\text{FeGe}_{0.25}\text{Si}_{0.75}$ ,  $\text{Co}_2\text{MnGa}_{0.25}\text{Ge}_{0.75}$ ,  $\text{Co}_2\text{Mn}_{0.5}\text{Fe}_{0.5}\text{Ga}$  などの幾つかの合金が高いスピンドル率を持つことが実験的に示されている。またこれらの合金ならびにそれらの薄膜の構造とスピンドル率の相関を調査し、スピンドル率が合金の  $L2_1$  規則度と明瞭な相関のあることを実験的に示した。本研究で見出された  $\text{Co}_2\text{FeGe}_{0.25}\text{Si}_{0.75}$  合金を強磁性層とした面直電流巨大磁気抵抗素子は極めて高い磁気抵抗変化を示すことも実証されており、本研究で見出された高スピンドル率強磁性合金はスピントロニクス分野に新たなスピンドル率電流源として注目されると期待され、その学術的価値は高いと判断される。

よって、著者は博士（工学）の学位を受けるに十分な資格を有するものと認める。