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論文内容要旨

【Introduction】

Spintronics, which introduces spins of electrons as an additional degree of freedom in electronics, has been creating widespread interests due to its large potential implication in creation of novel electronic devices. One of the most successful discoveries in the research field of spintronics is a large tunnel magnetoresistance (TMR) effect at room temperature (RT) in a magnetic tunnel junction (MTJ), consisting of FM_1 / I / FM_2 structure where FM_1 and FM_2 are ferromagnetic layers and I is an ultrathin insulator [1, 2]. The MTJ has been receiving increasing attention due to its possible technological applications such as a memory cell in a magnetic random access memory (MRAM).

On the other hand, spin-dependent resonant tunneling effect in the MTJ is one of fascinating spin-dependent phenomena. Due to the exchange splitting of up and down spin bands for ferromagnetic materials, we can expect an oscillation and enhancement of TMR effect as a function of an applied bias voltage in a spin-dependent resonant structure.

Studies of spin-dependent resonant tunneling effect have been made in many theoretical approaches and a few experimental trials so far. There are major two kinds of approaches. One is spin-dependent resonant tunneling effect in the MTJ with a nonmagnetic spacer layer [3-5], where spin-polarized QW states are formed in the nonmagnetic spacer layer by spin-dependent scattering. The other is spin-dependent resonant tunneling effect in a double barrier MTJ (DMTJ) [6], where QW states are formed in the middle ultrathin ferromagnetic layer for both spin bands.

【Purpose of this study】

In order to understand the fundamental physics of spin-dependent resonant tunneling effect and explore a probability of application to spintronics devices, two experimental trials have been done in this study.

First experiment is the investigation on spin-dependent resonant tunneling effect in the MTJ with an epitaxial Ru spacer layer. The previous experiments have been mainly focused on noble metals as a spacer layer. However, from the view point of the correlation between the spin-polarized QW formation and interlayer exchange coupling, Ru is a promising candidate as the spacer layer due to its large interlayer exchange coupling energy. The spacer thickness dependence and the applied bias voltage dependence on TMR effect were investigated systematically.

Second experiment is the first trial of investigation on spin-dependent resonant tunneling effect in fully epitaxial DMTJs. Coherent tunneling effect in a fully epitaxial MgO(001) barrier was applied to the spin-dependent resonant tunneling structure in this study.

【Experimental procedures】

Pseudo-spin-valve type MTJs, consisting of $Co_{90}Fe_{10}$ (1.2 nm) / Ru(t nm) / Al(1.1 nm)-Ox / $Co_{90}Fe_{10}$(3 nm) were deposited on single crystal MgO(110) substrates by using helicon sputtering system for the first experiment. On the other hand, pseudo-spin valve type DMTJs consisting of Fe (50 nm) / MgO (2 nm) / Fe (t nm) / MgO (2 nm) / Fe (20 nm) were deposited on MgO (001) substrates using molecular beam epitaxy (MBE) for the second experiment. In both cases, samples were patterned into junctions of $10\times10\ \mu m^2$ in size by successive microfabrication processes with a combination of electron-beam lithography and Ar ion-milling.

Both TMR and current-voltage (I-V) curves were measured using a conventional dc four-probe method. During the measurement, tunneling electrons flow from the bottom to the top electrode with $V > 0$.

【Experimental results】

1. Spin-dependent resonant tunneling effect in MTJs with a Ru spacer layer [7]

Epitaxial growth of the bottom h.c.p.$(10\bar{1}0)$ $Co_{90}Fe_{10}$ / Ru bilayer, which was confirmed by X-ray diffraction, was obtained in this experiment. Figure 1.1 shows the Ru thickness (0.3 nm $\leqq t \leqq$ 1.7 nm) dependence of the TMR ratio under constant bias voltage application of + 15 mV. Although the TMR ratio of 30 % is observed in the conventional type MTJ without the Ru spacer layer, the insertion of the 0.3 nm thick Ru layer decreases the TMR dramatically down to less than 1.5 %. However, the TMR ratio clearly oscillates and even changes its sign as a function of the Ru thickness. From the comparison with the oscillation observed in the interlayer exchange coupling of $Co_{90}Fe_{10}$ (15 nm) / Ru (t nm) / $Co_{90}Fe_{10}$ (5 nm) structure, both these oscillations are thought to originate from the same physical origin. Since the oscillation periods are in good agreement with the expected one from the stationary vectors in the band dispersion of h.c.p. Ru$(10\bar{1}0)$, the origin of these oscillations are attributed to spin-polarized QW formation in the ultrathin Ru layer.

Applied bias voltage dependence of the TMR ratio is shown in Fig. 1.2 (1.3 nm ≦ t ≦ 1.7 nm). The TMR ratio clearly oscillates with sign changes in both positive and negative bias directions for t ≧ 1.3 nm. These oscillations are interpreted by considering the contribution of spin-polarized QW states to conductance. It is inferred from the comparison between experimental results and a simple calculation using the band dispersion of Ru$[10\bar{1}0]$ (Σ band) that QW states are formed by Σ_5 or Σ_6 d band electrons and hybridize with the Σ_1 band which dominates the tunneling conductance.

This is the first observation of clear oscillations of TMR effect under the bias voltage application through the spin-polarized QW states created in the nonmagnetic spacer layer.

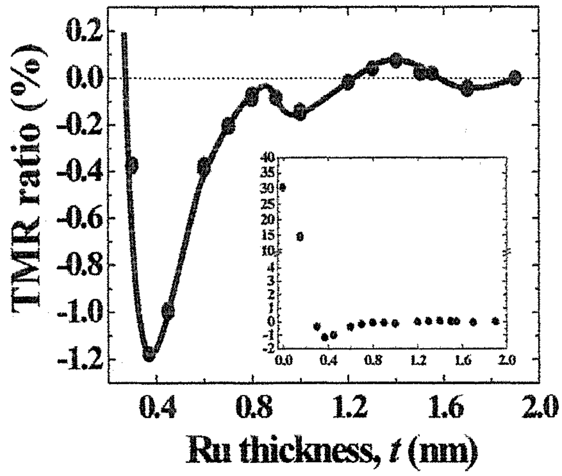

Fig.1.1 Ru thickness dependence of the TMR ratio in the Ru thickness range of 0.3 nm ≦ t ≦ 1.7 nm. The inset shows the entire Ru thickness dependence.

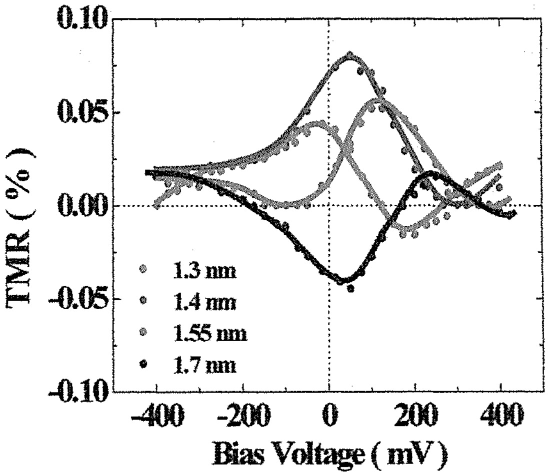

Fig.1.2 Bias voltage dependence of the TMR ratio in the Ru thickness range of 1.3 nm ≦ t ≦ 1.7 nm.

2. Spin-dependent resonant tunneling effect in fully epitaxial DMTJs [8, 9]

From RHEED patterns of each layer during deposition and after annealing treatment, fully epitaxial growth of Fe (001) / MgO (001) / ultrathin Fe (001) / MgO (001) / Fe (001) was confirmed. However, as shown in the cross-sectional TEM (Fig. 2.1, t = 1.5 nm), the middle Fe layer forms isolated islands (dotted area) of 20-60 nm in diameter. In addition, the actual thickness of the middle layer is increased due to aggregation. Since the tunneling current pass through the Fe islands selectively due to the thick barrier thickness besides the islands, the whole junction can be recognized as parallel-connected DMTJs.

The middle layer thickness dependence of conductance curves in the parallel magnetization configuration appears in Fig. 2.2. Clear oscillations of conductance are observed in the positive bias direction. Since the bias voltage where the conductance shows the maximum changes to a higher value with decreasing the middle layer thickness, the origin of these oscillations is thought to originate from the effect of QW states created in the middle Fe islands. The oscillatory featureobserved in the parallel magnetization state disappears completely in the antiparallel state. This result indicates that the QW states are formed by only one of two spin bands. From the simple consideration of the coherent tunneling mechanism in Fe(001) / MgO(001) / Fe(001) structure, we can speculate that these spin-polarized QW states are ascribed to the majority Δ_1 band states in the Fe (001) electrode.

The simple estimation of energy intervals of QW levels in an ideal double barrier tunnel junction was also performed using a transfer matrix method. The oscillation periods observed in the experiments were in good agreement with the calculation results when we assume the majority Δ_1 band of Fe(001) layer as confined electrons. However, the influence of an applied bias voltage on the oscillation period is not clear at this stage.

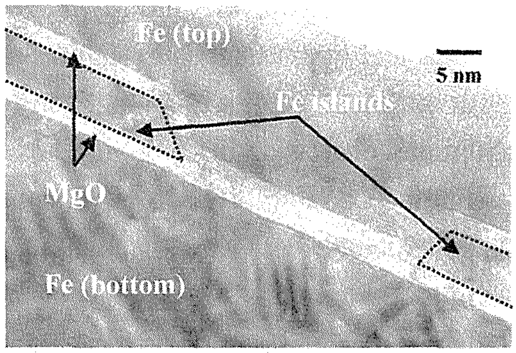

Fig.2.1 Cross-sectional TEM image of the DMTJ with the designed middle layer thickness of t = 1.5 nm.

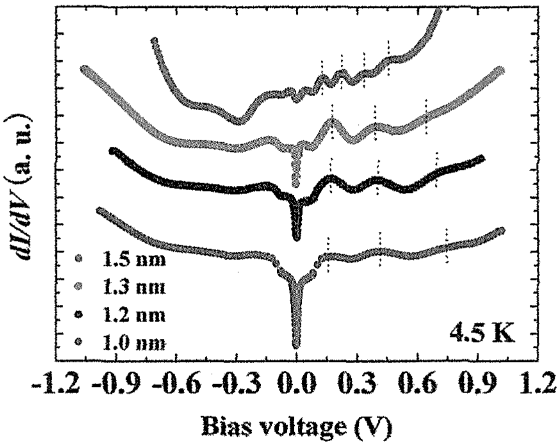

Fig.2.2 Middle layer thickness dependence of the conductance curves for the DMTJs in the parallel magnetization configuration.

【Conclusions】

Tunnel magnetoresistance (TMR) effect and conductance properties in magnetic tunnel junctions (MTJs) with an ultrathin nonmagnetic spacer layer and fully epitaxial double barrier magnetic tunnel junctions with an ultrathin middle ferromagnetic layer were investigated.

1. Clear oscillations of the TMR ratio as a function of the Ru spacer thickness were observed in the MTJs with a highly oriented $Ru(10\bar{1}0)$ spacer layer. The TMR ratio also oscillates as a function of the applied bias voltage with sign changes. These results can be explained by

considering the contribution of spin-polarized QW states created in the Ru spacer layer to conductance.

2. TMR and conductance properties of fully epitaxial DMTJs with layered Fe islands as a middle layer were investigated. Clear oscillations of conductance with the middle layer thickness dependence were observed only in the parallel magnetization state. The origin of these oscillations is attributed to the modulation of tunneling conductance by the spin-polarized QW states created in the middle Fe islands.

This is the first observation of the quantum size effect in the fully epitaxial DMTJs. However, the oscillations of resistance and TMR ratio as a function of the bias voltage are very small from the standpoint of applications. Further reduction of the middle layer thickness and the prevention of the islands formation are required for larger enhancement. These improvements of quality of the quantum well layer will provide the prominent QW effects and the realization of spin-dependent resonant tunneling effect.

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論文審査結果の要旨

強磁性トンネル接合(MTJ)は、室温で高いトンネル磁気抵抗(TMR)変化率を有することから、磁気抵抗効果型ランダムアクセスメモリ(MRAM)や磁気センサーなどへの応用を目指した研究が活発に行われている。著者は、MTJ 素子の機能性拡大を目指し、スピン依存共鳴トンネル効果の実現を目的とし本研究を行った。このため、超薄膜非磁性金属スペーサー層を有する MTJ 素子、および超薄膜中間強磁性層を有するエピタキシャル強磁性 2 重トンネル接合に関して、その TMR 特性の量子井戸層厚依存性、およびバイアス電圧の効果について詳細に調べた。本論文はこの研究成果についてまとめたものであり、全編 6 章よりなる。

第 1 章は序論であり、本研究の背景および目的を述べている。

第 2 章は、本研究成果を理解するために必要な TMR 効果の基礎的な特性と、最近の研究状況について概説している。第 3 章は、実験方法、および測定方法について述べている。

第 4 章では、非磁性スペーサー層を有する MTJ 素子の共鳴トンネル効果について述べている。著者は、層間交換結合の物理起源がスピン依存散乱による量子井戸形成であることに着目し、大きな層間交換結合エネルギーを示す Ru を量子井戸層に適用することを試みた。そのため、CoFe / Ru / AlOx / CoFe 構造からなる MTJ 素子を作製し、TMR 比の Ru 層厚依存性、およびバイアス電圧依存性について調べた。その結果、TMR 比が Ru 層厚に依存し、符号反転を伴う振動変化を示すことを明らかにした。観測された振動周期が層間交換結合の振動周期と類似することを確認し、これらの異なる 2 つの物理現象がともにスピン偏極量子井戸の形成に起因することを指摘した。また、バイアス電圧に対しても TMR 比が符号反転を伴いながら振動的に変化するという新しい物理現象の観測に成功した。Ru のバンド分散との対応から、これらの振動の起源となっている量子井戸の形成は Ru 中の d バンド電子であり、s 電子との混成により TMR 効果の振動現象として観測されたことを定性的に議論している。

第 5 章では、エピタキシャル Fe(001) / MgO(001) / Fe(001)構造の MTJ 素子におけるコヒーレントなトンネル効果、および高い TMR 効果に着目し、超薄膜中間層を有するエピタキシャル強磁性 2 重トンネル接合、Fe(001) / MgO(001) / ultrathin Fe(001) / MgO(001) / Fe(001)の作製を試み、その TMR 特性、およびコンダクタンス特性の中間層厚依存性を詳細に調べている。MgO と Fe の表面エネルギーの差によって、中間超薄膜 Fe 層は島状成長するが、成膜条件の最適化によって、エピタキシャル関係を保持する並列接続型の 2 重トンネル接合(DMTJ)を実現している。これらの DMTJ に関してコンダクタンス特性の中間層設計膜厚依存性を調べた結果、平行磁化配列状態において、コンダクタンスの振動現象の観測に成功した。観測された振動の周期が、設計膜厚の減少に伴って長周期へと変化していること、および反平行磁化配列状態においては、振動成分が完全に消失する点を考慮し、これらの振動の起源が中間 Fe(001)層中に形成された、majority Δ_1 バンド電子によるスピン偏極量子準位によるトンネルコンダクタンスの変調であることを指摘した。また、Transfer Matrix 法を用いた計算により、これらの議論の妥当性についても述べている。これは、2 重トンネル接合における初めての量子効果の観測である。

第 6 章は本論文を総括した結論である。

以上要するに本論文は、スピン依存共鳴トンネル効果という先駆的な研究に関して、TMR 効果の振動現象といった新しい物理現象の観測に成功しているだけでなく、その基礎物性に対する重要な知見を与えており、材料物性学の発展に寄与するところが少なくない。

よって，本論文は博士(工学)の学位論文として合格と認める。