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　２年　６月２３日現在

機関番号：８２６２６
研究種目：若手研究
研究期間：2018～2019
課題番号：１８Ｋ１３８０７

研究課題名（和文）Development of novel ferromagnetic tunnel contacts for efficient Si spintronic devices

研究課題名（英文）Development of novel ferromagnetic tunnel contacts for efficient Si spintronic devices

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交付決定額（研究期間全体）：（直接経費）　　3,200,000円

研究成果の概要（和文）：Siチャネル上のエピタキシャル強磁性(FM)トンネル接触の作製は,スピン金属-酸化物-半導体電界効果トランジスタのような半導体ベースのスピン輸送デバイスを開発する鍵である。このプロジェクトでは,新しいエピタキシャル酸化物トンネル障壁としてのSrO(001)のすることを検討しコヒーレントスピン偏極トンネリングが起こるかどうか。エピタキシャルSrOベースの磁気トンネル接合で大きな磁気抵抗比が達成できることを示し,スピン分極したコヒーレントた。SrOはMgOとSiの間よりもSiとの格子不整合が小さいので,この材料はSiベースの横方向スピン輸送デバイスにおいて高い磁気抵抗比を達成するための有望。

研究成果の学術的意義や社会的意義
半導体でのスピンの使用することにより,新しい機能を持つデバイスの開発が可能になる。
半導体におけるスピンの生成は、強磁性(FM)トンネルコンタクトからSCへの電気的スピン注入によって達成される。したがって、高スピン分極FMトンネルコンタクトの選択は、半導体において大きなスピン分極を生成するための重要なパラメータである。MgOはSiとの大きな格子不整合にもかかわらず,Si上のエピタキシャルトンネル障壁として今日までもっぱら使用されてきた。このプロジェクトでは,新しいトンネル障壁としてのSrOの利用を探求し,それが効率的なSiベースのスピン輸送デバイスを開発するための有望な材料であることを示した。

研究成果の概要（英文）：The fabrication of an epitaxial ferromagnetic (FM) tunnel contact on a Si channel is the key to developing semiconductor-based spin-transport devices such as a spin metal-oxide-semiconductor field-effect transistor. In this project, we explore the use of a novel epitaxial oxide, SrO(001), as a tunnel barrier and investigate whether coherent spin-polarized tunneling occurs through the SrO(001) tunnel barrier. We demonstrate that large magnetoresistance ratios can be achieved in epitaxial SrO-based magnetic tunnel junctions, indicating spin-polarized coherent tunneling. Since SrO has a smaller lattice mismatch with Si (5%) than that between MgO and Si (23%), this material appears as a promising epitaxial tunnel barrier for achieving high magnetoresistance ratio in Si-based lateral spin transport devices.

研究分野：Spintronic devices

キーワード：tunnel barrier　magnetic tunnel contact　magnetoresistance ratio　epitaxial growth

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2年6月10日現在

1. 研究開始当初の背景(Background)

The fabrication of an epitaxial ferromagnetic (FM) tunnel contact on a Si channel is the key to developing semiconductor-based spin-transport devices such as a spin metal-oxide-semiconductor field-effect transistor (spin-MOSFET). To date, MgO has been exclusively used as an epitaxial tunnel barrier on Si.[1-6] Recently we have succeeded in injecting a highly spin-polarized current ($P \sim 0.9$ at 10 K; P is the spin polarization of tunneling electrons) from epitaxial Fe(001)/MgO(001) tunnel contacts into Si(001) channel.[5-6] Such high P could, in theory, give a magnetoresistance ratio (MR) of several hundred %, according to the standard spin-transport model for a lateral device consisting of semiconductor channel with two FM contacts, like a spin-MOSFET.[7] However, such high P has only been achieved in a very high resistance-area products (RA) region (> 1 M$\Omega\mu m^2$) for Fe/MgO contact,[5-6] which significantly suppresses the MR.[7] In a low RA region (< 10 k$\Omega\mu m^2$), on the other hand, the tunneling electrons can no longer keep such high P (for example, P < 25% at 10 K).[5-6] The observed degradation of P in the low RA region can possibly be attributed to a poor crystalline quality of the very thin (< 1 nm) MgO tunnel barrier caused by the large $\Delta a/a$ between MgO and Si (22.6%).[8] Consequently, the MR so far achieved in Si-based lateral devices has been less than 1%.[1] Therefore, it is desirable to introduce a new tunnel barrier material which, from the standpoint of high-quality thin epitaxial tunnel barrier, can be adequately lattice-matched with Si, and thereby achieving high MR effect in Si-based lateral device.

2. 研究の目的(Purpose)

Owing to its relatively small lattice mismatch ($\Delta a/a$) with Si (4.97%) and its rock-salt structure, SrO(001) is possibly a suitable and efficient tunnel barrier for Si. Although the growth of a high-quality epitaxial of SrO(001) film on Si(001) has been demonstrated from an initial stage of the growth.[9] the application of SrO to a spin-transport device has been limited to an amorphous tunnel barrier on graphene.[10] In this project, we investigate the structural and magneto-transport properties of an epitaxial Fe/SrO/MgO/Fe magnetic tunnel junctions (MTJs) to determine whether coherent spin-polarized tunneling occurs through the SrO(001) tunnel barrier. The results of this project will in turn clarify whether SrO(001) is a promising candidate as a high-quality tunnel barrier for Si-based spin transport devices.

3. 研究の方法(Method)

MTJ films as presented in Fig. 1 were grown by molecular beam epitaxy (MBE). The films consisted of a Au cap (10 nm) / Co pinned layer (20 nm) / Fe top electrode (10 nm) / SrO tunnel barrier (1.4 nm) / MgO underlayer (0.8 nm) / Fe bottom electrode (30 nm) / MgO buffer layer (5 nm) on a MgO(001) substrate. The MgO underlayer enables the SrO tunnel barrier to grow epitaxially, and acts as an epitaxial tunnel barrier. The source materials were evaporated using electron-beam guns (for Fe, SrO, and MgO) and Knudsen-cells (for Au and Co). Single-crystal SrO granules and MgO block were used as source materials. Prior to the growth, the MgO substrates were cleaned by an ultrasonic cleaner with acetone and isopropanol, and thermally annealed at 800 °C for 10 min in the MBE chamber with a base pressure 2 × 10^{-9} Torr. The MgO buffer layer and Fe bottom electrode were deposited on the substrate at 100 °C, followed by an *in situ* annealing at 300 °C for 10 min to improve the surface morphology of the Fe bottom electrode. Then, the MgO underlayer was grown on the Fe bottom electrode at RT. Subsequently, the SrO tunnel barrier was deposited on the MgO layer at RT under an O_2 pressure of 1-3 × 10^{-7} Torr. The Fe upper electrode was grown on the SrO tunnel barrier at RT, and then annealed for 10 min at 300 °C to reduce the dislocation density at the Fe/SrO or SrO/MgO interfaces. Finally, Co-pinned and Au-cap layers were deposited onto the Fe top electrode at RT. As a reference, the same MTJ stack without the MgO underlayer was also grown.

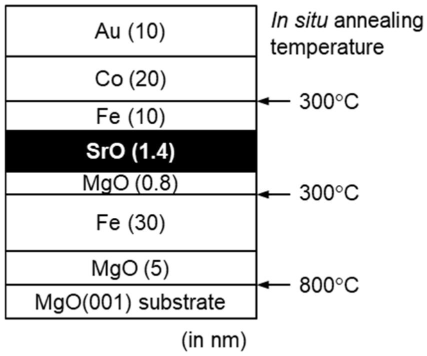

Fig. 1: Schematic structure of MTJ stack and *in situ* annealing temperature adopted during the deposition processes.

Structural properties of the MTJ films were investigated by *in situ* reflection high energy electron diffraction (RHEED), *ex-situ* cross-sectional high-angle annular dark field scanning transmission electron microscope (HAADF-STEM), and the elemental mapping by energy-dispersive x-ray spectroscopy (EDX). For the magneto-transport measurements, the films were patterned into tunnel junctions with active areas from 3 × 12 μm^2 to 6 × 24 μm^2 using conventional micro-fabrication techniques (e.g., photolithography, Ar ion milling, and SiO_2 sputtering). The measurements were carried out using a conventional DC two-probe method. The magnetic fields were applied parallel to the major axis of the junction corresponding to the easy axis of the magnetization direction of the Fe electrodes.

4. 研究成果(Results)

4.1. Structural characterizations:

Structural analyses were performed on the fully epitaxial film by means of HAADF-STEM and EDX observations as illustrated in Figs 2(a) and 2(b), respectively. Both images revealed steep interfaces without interdiffusion among each layer. Note that, from the HAASF-STEM image, misfit dislocations in every several atoms at the Fe/SrO and SrO/MgO interfaces were clearly

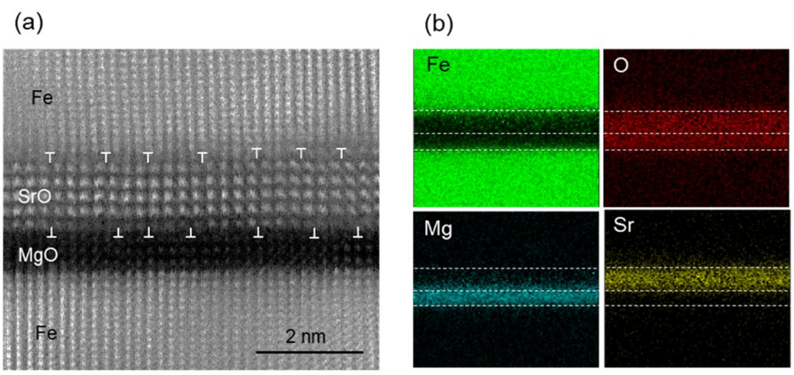

Fig. 2: (a) Cross-sectional HAADF-STEM image, and (b) EDX elemental mappings of Fe, O, Mg, and Sr of the Fe/SrO/MgO/Fe stack ([100] azimuth of MgO substrate).

observed, reflecting from the effect of large $\Delta a/a$. By assuming a 45° in-plane rotation between the Fe and SrO layers and a cube-on-cube relation between the SrO and MgO layers, the in-plane $\Delta a/a$ between the SrO tunnel barrier and Fe top electrode, and that between the SrO tunnel barrier and bottom electrode are both estimated to be -21.6%. The estimated value is very close to the expected value from the bulk material (-21.4%). Accordingly, the in-plane crystal orientations were determined as top Fe[110] || SrO[001] || MgO[001] || bottom Fe[110], respectively. Note that the 45° in-plane rotation between the MgO(001) tunnel barrier and Fe(001) electrodes is a preferable crystal orientation for the coherent spin-polarized tunneling.[1-3] Since SrO has the same crystal structure as MgO, the result implies that we can expect coherent tunneling with this type of MTJ.

4.2. Magneto-transport properties

Typical magnetoresistance (MR) curves of the epitaxial MTJ are shown in Fig. 3(a). Here, the MR ratio is defined as $(R_{AP} - R_P)/R_P$, where R_P and R_{AP} are the junction resistances in parallel (P) and antiparallel (AP) magnetization states, respectively. We observed MR ratios up to 98% at 20 K and 65% at RT, respectively. The observed MR ratios are almost twice as high as those reported in a polycrystalline Fe/amorphous GaO_x/MgO(001)/Fe(001) MTJ (50% at 20 K and 34% at RT, respectively),[11] where the coherent spin-polarized tunneling is considerably suppressed due to the poly-crystalline nature of the Fe top electrode and amorphous GaO_x tunnel barrier. This strongly suggests that coherent spin-polarized tunneling occurs through the SrO(001) tunnel barrier, and Δ_1 state acts as the major tunneling channel in the P state.[12-15] In addition, it was found that the R_P has small temperature dependence, namely, relative changes in the R_P between 20 K and RT were very small (2-4%) compared with those for the R_{AP} (10-13%). This is a typical feature of fully epitaxial MgO- and $MgAl_2O_4$-based MTJs,[15,16] in which coherent tunneling has been both experimentally and theoretically demonstrated. The result supports our conclusion that the coherent tunneling is achieved in the epitaxial Fe/SrO/MgO/Fe MTJ.

We did not observe MR effect nor a non-linear (tunnel-like) current-voltage characteristics in the reference MTJ, implying that there are many imperfections such as pinholes in the polycrystalline SrO tunnel barrier.

It is interesting to know how the asymmetric barrier/electrode structure having such high-density dislocations among the interfaces affects the spin-dependent tunneling. In general, bias-voltage (V) dependence of the MR ratio is sensitive to the barrier and barrier/electrode interface qualities. Namely, poor barrier quality results in a low bias-V at which the MR ratio reaches half of the zero-bias value (V_{half}). As a result, rapid decrease in the MR ratio often occurs in a bias-V direction where the electrons tunnel into the electrode having a lower interface quality. In Fig. 4(b), the MR ratio at RT is

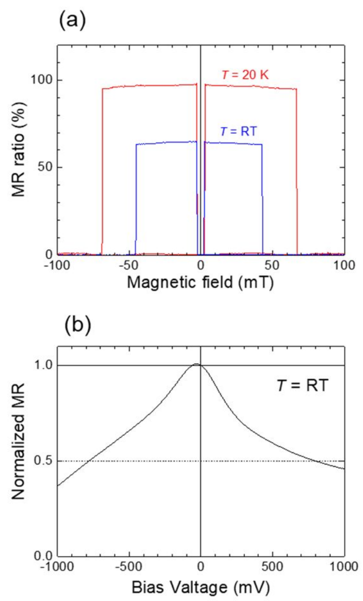

Fig. 3: (a) Magnetoresistance curves of the Fe/SrO/MgO/Fe MTJ at 20 K and RT applying 10 mV voltage, and (b) bias-voltage (V) dependence of the normalized MR

plotted as a function of bias-V for the epitaxial Fe/SrO/MgO/Fe MTJ. Here, positive bias is defined as the bias direction where the electrons tunnel from the Fe bottom electrode (MgO/Fe interface) into the Fe top electrode (Fe/SrO interface). The plot was almost symmetric despite the asymmetric structure, and the V_{half} became as high as 800 mV regardless of the bias directions. The observed high V_{half} is even comparable to those in lattice-matched systems of epitaxial Fe/MgO/Fe,[14] Fe/$MgAl_2O_4$/Fe[16] and Fe/GaO_x/MgO/Fe[11] MTJs. Consequently, we could not observe clear evidence of the influence on the MR ratio for the asymmetric barrier/electrode structure with many dislocations at the interfaces. Conversely however, the observed high V_{half} in both bias directions are favorable for efficient spin-injection/detection schemes in a Si-based lateral device with two FM contacts.

To conclude, we studied the structural and magneto-transport properties of the lattice-mismatched of Fe/SrO/MgO/Fe MTJ. The structural analyses revealed a fully epitaxial Fe(001)/SrO(001)/MgO(001)/Fe(001) structure having many misfit dislocations at the Fe/SrO interface. Despite the existence of such misfit dislocations, high MR ratio up to 98% at 20 K (65% at RT) was observed, indicating that coherent spin-polarized tunneling takes place through the SrO(001) tunnel barrier. The V_{half} values in both bias directions were as high as 800 mV, which are comparable to the reported value in the epitaxial Fe/MgO/Fe MTJs. Since SrO has a much smaller $\Delta a/a$ with Si, SrO(001) appears as a promising epitaxial tunnel barrier for achieving high MR ratio in Si-based lateral spin transport devices.

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５．主な発表論文等

〔雑誌論文〕　計0件

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４．発表年 2019年

〔図書〕　計0件

〔産業財産権〕

〔その他〕

A manuscript entitled: "Structural and magneto-transport properties of lattice-mismatched epitaxial Fe/SrO/MgO/Fe magnetic tunnel junctions" has been submitted to Japanese Journal of Applied Physics (JJAP) in June, 2020.

６．研究組織

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