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論 文 内 容 要 旨

Background (Chapter 1)

Femto-second laser pulse induces a transient spin current in ferromagnetic metal thin films. The laser pulse-induced spin current attracted much attention for technological points of view, such as a way to excite magnetization dynamics in sub-ps time scale, and terahertz (THz) pulsed electromagnetic wave emission. An efficient spin current generation in ultrathin metal films composed of ferromagnetic metal (FM)/heavy metal (HM) bilayer was reported in 2016 by T. Seifert *et al*. Although several demonstrations and analysis based on the physical model of spin current generation process in thick ferromagnetic metal films have been reported, there are no systematic studies on laser pulse-induced spin current in ultrathin films and its mechanism is not clear. Hence; an investigation of ferromagnetic metal layer dependence of the laser pulse-induced spin current in ultrathin films is required. Ta/CoFeB/MgO multilayer is a well-studied structure and its stability for ultrathin regime have been reproduced by many groups. Especially for the annealing effect and crystallization process, the MgO capping layer acts as a template for a (001) orientation of CoFeB layer to form a bcc CoFe. Its magnetic properties are drastically changed by choosing its composition and annealing temperature as reported in giant magnetoresistance (GMR) and tunnel magneto resistance (TMR) studies. Thus, further detailed research on composition and annealing temperature dependence is required to get insight into the detailed feature of laser pulse-induced spin current. Purposes of this thesis are given as follows; 1) To construct new all-optical set up for the laser pulse -induced spin current measurement with an electromagnet and magnetooptical Kerr effect measurement set up. 2) To investigate the laser pulse induced-spin current in Ta/CoFeB/MgO films and to construct a phenomenological model based on experimental results in order to get insight into a relationship between the laser pulse-induced spin current and the effects of crystallization, saturation magnetization, and spin polarization of ferromagnetic metals. 3) To clarify the carrier and transport property of the pulse laser-induced spin current.

Experimental Procedures (Chapter 3 and Section 4.1)

Ta(5.0)/$(Co_xFe_{1-x})_{80}B_{20}$($t_{CFB}$)/MgO(2.0)/Ta(2.0) (thickness in nm) multilayer films were deposited on thermally oxidized Si substrates using ultrahigh vacuum magnetron sputtering. The samples were annealed at 150-500°C in vacuum furnace. The film thickness t_{CFB} was varied from 0.4 to 10.0 nm. In order to investigate the laser pulse-induced spin current, a THz time-domain spectroscopy (THz-TDS) was performed by means of an electrooptic sampling with a Ti: sapphire laser and regenerative amplifier (a pulse duration of about 120 fs). The THz emission mechanism proposed was as follows: the laser pulse-induced spin current flows from the CoFeB layer into the Ta layer induces transverse electric current in Ta layer due to the inverse spin-Hall effect, and it emits a pulsed-THz electromagnetic wave to air. By measuring an intensity of the THz wave emitted in time-domain with pump-probe method, the laser pulse-induced spin current in sub-ps time scale was investigated. In this study, an optical set up for THz-TDS with a sensitivity of 18 V/cm, a THz frequency range of 0.1~3.0 THz, and a maximum applied magnetic field of 2 T was constructed.

Experimental Results (Chapter 4)

Section 4.2: CoFeB thickness and annealing temperature dependence

In order to discuss the laser pulse-induced spin current, the CoFeB thickness and annealing temperature dependence of THz emission intensity were investigated in Ta/$Co_{20}Fe_{60}B_{20}$/MgO multilayered films. A 1.5-fold increase in the THz wave emission intensity after the annealing at T_a = 300 °C was observed for the CoFeB layer thickness of 1.0-2.0 nm. The thickness dependence of the THz wave emission intensity also changed with increasing annealing temperature. However, there is no annealing temperature dependence of the ultrafast demagnetization value considered partially related to each other. Similar changes in the saturation magnetization, conductivity and THz wave emission intensity were observed. These results indicate the effect of the crystallization of CoFeB on the THz emission intensity. The spin-dependent mean free path $\lambda_{\uparrow}$ ($\lambda_{\downarrow}$) enhancement after the crystallization of CoFeB layer would be quantitatively consistent with the enhancement of the THz wave emission intensity for the films with the 4 to 10-nm-thick CoFeB layer.

Section 4.3: MgO barrier thickness dependences

To get insight of the MgO insertion effect on the THz wave emission intensity, the annealing temperature T_a dependence of THz wave signal in Ta/$Co_{20}Fe_{60}B_{20}$ (t_{CFB})/Ta films were investigated. Although the THz wave signal slightly increased after the annealing at T_a = 500 °C in t_{CFB} = 10 nm film, no huge enhancement was observed, unlike the results with MgO insertion layer as described in Section 4.2. To estimate the spin current flow into the cap heavy metal layer through the MgO layer, MgO layer thickness dependence of THz wave emission intensity in Ta(3.0)/ $Co_{20}Fe_{60}B_{20}$(2.0) /MgO(t_{MgO})/Pt(2.0) (thickness in nm) was investigated. Because the top Pt capping layer has a large spin Hall angle, the THz wave emission intensity increased in the films without MgO insertion layer which is expected to cut off the spin current flow into the Pt layer. However, the THz wave emission intensity exponentially decreased with increasing t_{MgO}. It is clarified that the MgO insertion layer with a thickness of 1.0 nm completely cut off the spin current flow to the top heavy metal layer.

Section 4.4: Ta buffer layer thickness dependence

In order to discuss the annealing effect on the THz emission, the sample with various thickness of Ta buffer layer were deposited. The stacking structures were Ta(t_{Ta})/$Co_{20}Fe_{60}B_{20}$(2.0)/MgO(2.0)/Ta(2.0) (thickness in nm). These samples were annealed at 200, 300, and 400°C for 1 h in vacuum. Although the temperature dependence of THz emission intensity depends on Ta buffer layer thickness due to the surface structure or something features which depends on the buffer layer, the THz emission intensity at optimized annealing temperature increased 1.8 times from the intensity of as-deposited films; that increment was independent of the Ta buffer layer thickness. Spin Hall angle depends on impurity or resistivity of the heavy metal layer. Boron in CoFeB layer is well-known that boron diffuses into the Ta buffer layer during the annealing process and crystallization. When we consider the boron diffusion effect on spin Hall angle in Ta due to the increase in impurity density, the enhancement of THz emission intensity after the annealing process should be higher in thinner Ta buffer films, as the density of boron or impurity would be higher. Thus, the experimental results indicate that no boron impurity effected on the Hall angle of Ta and no boron (impurity) density dependence of THz emission intensity was observed in this study.

Section 4.5: Composition dependence of THz emission in Ta/$(Co_xFe_{1-x})_{80}B_{20}$/MgO films

Samples were fabricated by using co-sputtering method with $Fe_{80}B_{20}$ and $Co_{80}B_{20}$ targets. The Co composition *x* was varied from 0.0 to 0.75. Although no composition dependence was observed in as-deposited CoFeB films, as seen in previous report by T. Seifert, *et al*, the clear and significant composition dependence was observed in the crystalized CoFeB films in this study. The THz emission intensity exhibited a maximum at the Co composition of *x*=0.1~0.3 in the films with the CoFeB thickness of 4.0 nm. The composition dependence of the THz emission intensity slightly changed in the films with the CoFeB thickness of 1.4 and 2.0 nm. The ultrafast demagnetization value and sheet conductivity slightly increased with increasing Co composition *x*. The trend of the composition dependence of the saturation magnetization is qualitatively similar with that of the laser pulse-induced spin current intensity in all films. However, it is difficult to quantitatively discuss the composition dependence by comparing with the saturation magnetization.

Section 4.6: Experiments using ultra-short optical pulse

To discuss the change in the THz waveform via an ultra-short laser pulse with a pulse duration of 10 fs, THz wave emission measurements by using ultra-short optical pulse were performed in Fritz Harber Institute, Berlin. When the FeB layer thickness decreased to 1.0 nm from 4.0 nm, THz wave waveform and Fourier spectrum showed similar waveform and spectrum with the signal after the annealing process. Although the experimental results showed systematic trend, there is no idea to discuss. Further investigation of the wave form change due to the thickness and normal metal layer is required.

Discussion (Chapter 5)

Based on a ballistic electron transport model, the spin current induced by the pulse laser was calculated. The thickness dependence of the THz wave emission intensity was converted into the laser pulse-induced spin current intensity by considering the impedance matching. Because the optical penetration depth is much larger than the films thickness, the ballistic carrier is excited in whole metal layer and transports thickness direction. Since the spin polarized carrier is excited in ferromagnetic metal layer and the spin current , which induces THz local field, flows from ferromagnetic metal layer to heavy metal layer, the excited carrier transports from ferromagnetic metal layer to heavy metal layer was considered. The thickness dependence of the laser pulse-induced spin current intensity in the amorphous and crystalized CoFeB films can be explained by using the spin-dependent mean free path of $\lambda_{\uparrow}$=5, 1.6, 0.8 and $\lambda_{\downarrow}$=1.0, 0.5, 0.4 nm for the films annealed at 400, 300 and as deposited films, respectively. These values are similar to the values reported using a DC transport measurement technique. To discuss the Co composition dependence of the laser pulse-induced spin current intensity and compare with the saturation magnetization and band structure, optical excitation was considered. Because of the optical selection rules, the optical excitation from *p*(*d*) band to *d*(*p*) band was considered. The joint density of states for an optical transition from 3*d* to 4*p* state in the majority spin band and from 4*p* to 3*d* state in the minority spin band were numerically calculated as a spin polarization of the optically-excited electron-hole carrier. The evaluated spin polarization remarkably decreased with increasing the Co composition in CoFe. This trend was quantitatively consistent with the Co composition dependence of the laser pulse-induced spin current intensity. Though the trend is consistent with experimental results, the increase of the THz wave emission intensity around Co composition of x = 0.1–0.3 in t_{CFB} = 4.0 nm films is still open. Further consideration is required by concerning the transport property or band transition.

Conclusion (Chapter 6)

The THz wave emission measurement set up was constructed for the laser pulse-induced spin current experiments. The laser pulse-induced THz wave emission in the Ta/$(Co_xFe_{1-x})_{80}B_{20}$/MgO/Ta films with various CoFeB layer thicknesses t_{CFB}, annealing temperatures T_{a}, and Co composition *x* were discussed to investigate the correlations between the laser pulse-induced spin current and the saturation magnetization, conductivity or spin polarization of optically excited carrier in the crystallized CoFeB layer. These experimental results were explained using the phenomenological model based on the ballistic electron transport and the optical excitation of the electron-hole carrier. These results strongly contribute to development of the physics for the laser pulse-induced spin current and the technology for the ultrafast spin control.

論文審査結果の要旨

第1章は序論である。金属強磁性薄膜におけるパルスレーザー誘起スピン流、そしてスピン流がテラヘルツ（THz）波を放射する現象について先行研究も交えて説明し、本研究の位置づけを述べている。また、Ta/CoFeB/MgO 多層膜の基本的な物性について述べた後に、本研究の目的について述べている。

第2章は実験原理である。スピン流を記述する拡散方程式、スピン流から電流への変換原理、パルス電流から発生する THz 波、金属薄膜から THz 波が放射される際の放射効率とインピーダンス整合の関係について述べている。

第3章は実験方法である。Ta/CoFeB/MgO 多層膜試料はマグネトロンスパッタ法を用いて製膜した。基板には熱酸化膜付き Si 基板を用いた。光源として、チタンサファイアパルスレーザーおよび再生増幅器を用いた。THz 波放射の測定にはポンプ・プローブ法と電気光学サンプリングによる THz 時間領域分光法を用いた。

第4章は実験結果である。まず、本研究で構築した光学系について述べている。最大印加磁場が 2 T 印加できる光学系を構築した。次に、CoFeB 膜厚及び熱処理温度の異なる多層膜試料に関する実験結果を述べている。THz 波放射強度は試料熱処理によって 1.5 倍程度増大することが分かった。また、THz 波放射強度が CoFeB 膜厚に強く依存すること、その依存性が試料の熱処理温度によって大きく変化することが分かった。次に、THz 波放射強度の MgO および Ta 層厚依存性に関する実験結果を述べている。THz 波放射強度は MgO 層膜に依存するものの、1 nm 以上ではほぼ変化がないことが分かった。また、熱処理による THz 波放射強度の増大は Ta 膜厚に依存しないことが分かった。さらに、THz 波放射強度の CoFeB 組成比依存性について述べている。熱処理後の試料において THz 波放射強度の Co 組成比依存性は顕著であり Co と Fe の組成比が約 3:7 付近で最大をとることが分かった。最後に、フリッツハーバー研究所（ベルリン）で行われた高帯域の評価に関して述べている。熱処理後に THz 波の波形がわずかに変化することが分かった。フーリエ解析の結果、0.1-5.0 THz 付近の周波数成分が増大し実時間波形が変化していることが分かった。

第5章は実験結果の考察である。光吸収による電子・ホール対の励起とその励起キャリアの弾道的な輸送のみをスピン流の起源と仮定したモデルを構築した。励起電子・ホール対のスピン密度を連結状態密度から求め、その Co 組成比に対する変化をリジットバンドモデルから評価した。THz 波放射強度の Co 組成比依存性は、p バンドのみを考慮した励起電子・ホール対のスピン密度の組成依存性で定性的に説明できることが分かった。また、THz 波放射強度の CoFeB 膜厚依存性や熱処理の効果は、励起スピン密度やスピンに依存した平均自由行程を考慮することで説明できることが分かった。これらの考察により、極薄膜におけるパルスレーザー誘起スピン流は、光励起遷移によって生じる非平衡スピン密度とその弾道的な輸送が一つの起源であることが示唆された。

第6章は総括ならびに今後の展望である。Ta/CoFeB/MgO 構造において THz 波検出を通してパルスレーザー誘起スピン流を調べた。熱処理によって CoFeB が結晶化するとその膜厚依存性や組成比依存性が大きく変化することが分かった。これはパルスレーザー誘起スピン流そのものが大きく変化していることを意味している。実験結果を簡易なモデルで考察し、光誘起スピン流が光励起遷移によって生じる非平衡スピン密度とその弾道的な輸送を一つの起源とすることが明らかとなった。今後第一原理計算を援用することでさらに明確な理解が得られると期待される。以上、本研究で得られた知見は、スピン流を用いたデバイスの基礎的な評価手法を確立していくうえで非常に有用なものであり、スピントロニクスの発展、ひいては応用物理学の発展に大きく寄与すると期待される。

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。