論 文 内 容 要 旨

(NO. 1)

| 氏 名 | 日置 友智 | 提出年 | 令和2年 |
|--------------|--|--------------------------------|------------------------|
| 学位論文の 題 目 | A study on propagation dynamics of magnon-phonon using time-resolved magneto-optical i (時間分解磁気光学イメージングを用いたマグノン―フォノン港 | n hybridiz maging 記成波ダイナ | ed waves by ミクスの研究) |

論 文 目 次

| 1 | Intr | oduc | ction | 1 |
|---|------|--------|---|----|
| | 1.1 | Pret | face: Spintronics, spin wave, and magnonics | 1 |
| | 1.1 | .1 | Magnetization dynamics and precession | 2 |
| | 1.1 | .2 | Magneto-static wave | 4 |
| | 1.1 | .3 | Quantum theory of spin waves: magnons | 6 |
| | 1.2 | Nor | n-linear dynamics of magnetization | 8 |
| | 1.2 | .1 | Anharmonic precession of magnetization | 9 |
| | 1.2 | .2 | Parametric excitation of magnons | 10 |
| | 1.2 | .3 | Multi-magnon scattering | 14 |
| | 1.3 | Mag | gnon-phonon coupling | 15 |
| | 1.3 | .1 | Macroscopic model of magneto-elastic coupling | 16 |
| | 1.3 | .2 | Quantum theory of magneto-elastic coupling | 17 |
| | 1.3 | .3 | Spin-lattice relaxation | 20 |
| | 1.3 | .4 | Hybridization of magnon and phonon | 22 |
| | 1.4 | Pur | pose of this thesis | 26 |
| | | | | |
| 2 | San | nple j | preparation and measurement methods | 29 |
| | 2.1 | Pro | perties of $Lu_2Bi_1Fe_{3.4}Ga_{1.6}O_{12}$ | 29 |
| | 2.1 | .1 | Crystallographic structures | 30 |
| | 2.1 | .2 | Magneto-optical properties | 32 |
| | 2.2 | Fen | nto-second pulse laser system | 35 |
| | 2.2 | .1 | Parametric amplification of light | 35 |
| | 2.2 | .2 | Mode locking | 40 |
| | 2.2 | .3 | Pulse compression | 43 |

| | 2.2 | 2.4 | Regenerative amplifier | 45 |
|-----|-----|----------|---|----|
| | 2.3 | Tin | ne-resolved magneto-optical imaging: Spin wave tomography | 46 |
| | 2.3 | .1 | Optical setup | 46 |
| | 2.3 | 3.2 | Fourier analysis of magneto-optical images | 50 |
| 3 | TR | MO | imaging of magnetoelastic waves by Cotton-Mouton effect | 52 |
| | 3.1 | Ma | gnetization configuration and magneto-optical effect | 53 |
| | 3.2 | Exp | erimental method to obtain spin dynamics via Cotton-Mouton effect | 54 |
| | 3.2 | 2.1 | Setup for magneto-optical imaging by Cotton-Mouton effect | 54 |
| | 3.2 | 2.2 | Evaluation of stokes parameters of transmitted light | 55 |
| | 3.3 | Res | ults of imaging by the Cotton-Mounton effect | 56 |
| | 3.3 | .1 | Static imaging | 56 |
| | 3.3 | 3.2 | Time-resolved imaging | 58 |
| | 3.4 | Ma | gnetic field dependence of excitation intensity of magneto-elastic waves | 60 |
| | 3.5 | Sun | nmary | 62 |
| 4 | Sne | ell's la | aw for spin waves at a 90° magnetic domain wall | 63 |
| | 4.1 | Ref | lection and refraction law of spin waves | 63 |
| | 4.2 | Spir | n-wave refraction and reflection at a magnetic domain wall | 65 |
| | 4.3 | Inci | ident angle dependence of spin-wave refraction at a magnetic domain wall | 67 |
| | 4.4 | Sun | nmary | 70 |
| 5 | Bi- | reflee | ction of spin waves due to magneto-elastic mode conversion | 73 |
| 5.1 | 1 B | Birefr | ingence of a light and mode degree of freedom of a spin wave | 73 |
| | 5.2 | Exp | perimental methods | 75 |
| | 5.2 | 2.1 | Material characterization | 75 |
| | 5.2 | 2.2 | Magneto-optical imaging | 76 |
| | 5.3 | Ma | gneto-optical imaging of bi-reflection of spin waves | 77 |
| | 5.4 | SW | aT analysis of bi-reflection of spin wayes | 79 |
| | 5.4 | .1 | Magnetization orientation dependence of bi-reflection of spin waves | 79 |
| | 5.5 | Exn | perimental confirmation of avoided crossing due to magneto-elastic coupling | 80 |
| | 5.6 | Phe | enomenological modeling of magneto-elastic mode conversion | |
| | 5.6 | 5.1 | Incident angle dependence of bi-reflection of spin waves | |
| | 5.7 | Sun | nmary | 90 |
| | | | | |

| 6 | Col | Coherent oscillation of magnon and phonon | |
|----|--|---|-----|
| | 6.1 Coherent oscillation of magnon-phonon hybridized state | | |
| | 6.2 | Long-delay magneto-optical imaging of magneto-elastic waves | 92 |
| | 6.3 | Real space images of spin-wave dynamics | 94 |
| | 6.4 | SWaT analysis of coherent oscillation of magnon-phonon hybridized state | 95 |
| | 6.4 | .1 Spectrum analysis of the excited spin waves | 95 |
| | 6.4 | .2 Time domain analysis of amplitude of hybridized state | 97 |
| | 6.4 | .3 Magnetic field dependence of oscillation frequency | 98 |
| | 6.5 | Summary | 100 |
| 7 | Vid | eo frame interpolation for spin-wave videos by means of machine learning | 103 |
| | 7.1 | Interpolation of video as a tool for spin-wave spectroscopy | 103 |
| | 7.2 | Related method | 104 |
| | 7.3 | Our proposal | 105 |
| | 7.4 | Experiment and results | 108 |
| | 7.4 | .1 Synthesis of wave video | 108 |
| | 7.4 | .2 Evaluation of the interpolation results | 109 |
| | 7.5 | Discussion | 110 |
| | 7.6 | Conclusion | 110 |
| 8 | Сот | nclusion | 111 |
| 9 | Bib | liography | 114 |
| 1(|) Ack | xnowledgement | 122 |
| 11 | l List | t of publications | 126 |
| | 11.1 | Peer-reviewed papers | 126 |
| A | Арр | endix. 1: Calculation on the response of magnetic part of magneto-elastic waves | 128 |
| | A.1 | Derivation of equation of motion for magnetization in a perpendicularly magnetized film | 128 |
| | A.2 | Derivation of spin precession amplitude due to magneto-elastic coupling | 129 |

| В | Appe | endix. 2: Calculation of the dispersion relation of magneto-static waves in a magnetic film with | |
|-----|---------|--|------|
| arł | oitrary | magnetization orientation and magnetocrystalline anisotropy | 131 |
| | B.1 | Calculation of the dispersion relation of spin waves in a magnetic film with arbitrary | |
| I | magne | etization orientation | .131 |

In the field of spintronics, interaction between spin and lattice has been of interest because lattice has been regarded as a relaxation channel which determine the propagation length of a spin current, a flow of spin angular momentum. Within the framework of spin transport phenomena, the interaction between spin and lattice is understood by incoherent scattering between elementary excitations such as magnons and phonons, and there are few such phenomena that roots from the coherent interaction between the excitations. The magneto-elastic hybridization refers to the creation of new normal state consisting of magnon and phonon as a result of resonant and coherent coupling. The created hybridized state holds characteristics of both magnon and phonon, and thus expected to exhibit unconventional propagation dynamics.

This thesis addresses the propagation dynamics of magnon-phonon hybridized waves by using time-resolved magneto-optical imaging. The central purpose of this thesis can be divided into (i) establishing a magneto-optical imaging method for perpendicularly magnetized film (Chap. 3) and (ii) unraveling the refraction and reflection law of magnon-phonon hybridized waves (Chap. 4 and 5), and (iii) experimentally demonstrating coherent interexchange of magnon and phonon by magneto-elastic coupling and the trial to accelerate the experiment by machine learning (Chap. 6 and 7).

 Chapter 1 shows a brief history of spintronics and magnonics and explains basic concepts and studies which are necessary to understand the following Chapters. Besides, pioneering works and some topics that have attracted much attention in spintronics and magnonics are also introduced.
Chapter 2 describes the sample characterization and measurement methods. Crystallographic and magneto-optical properties of Lu₂Bi₁Fe_{3.6}Ga_{1.4}O₁₂ (LuIG) and working principles of pulse laser system used for the time-resolved magneto-optical imaging is also introduced in this Chapter.

• *Chapter 3* shows the magneto-optical imaging technique to visualize the magnetization dynamics in an out-of-plane magnetized film.

Direct observation of spin waves in a magnetic material has been expanding the field of magnonics where the propagation of spin waves plays a main role for information processing and transfer. In the physics point of view, spatio-temporal observation of spin wave propagation provides rich information on the magnon-magnon interaction and magnon-phonon interaction. Previous magneto-optical imaging technique utilizes Faraday effect, a magneto-optical effect proportional to the magnetization component along the propagation axis of the light, and thus magnetization dynamics in an out-of-plane magnetized film was not accessible. We demonstrated the time-resolved magneto-optical imaging by using Cotton-Mouton effect, a magneto-optical effect proportional to the magnetization component perpendicular to the propagation axis of the light. The technique allows us to observe the propagation dynamics of magneto-elastic waves in an out-of-plane magnetized sample, revealing the contribution of magneto-elastic coupling for the excitation of the wave. • *Chapter 4* shows the refraction law of spin waves in the presence of 90° magnetic domain wall. Spin waves are categorized into two types depending on the dominant interaction between local magnetizations. In the long wavelength regime, the interaction dominantly arises from magnetic dipole interaction, which breaks rotational symmetry. Thus, the dispersion relation of spin waves in the long wavelength regime is highly anisotropic depending on the orientation of the saturation magnetization. This leads to the abnormal reflection and refraction law at a magnetic boundary. We investigated spin-wave dynamics excited by a magneto-elastic coupling in the vicinity of 90-degree magnetic domain wall. We observed abnormal negative refraction of spin waves, and we modeled the law to formulate spin-wave version of Snell's law. The result provides an approach to realize the cloaking of object from spin waves as tried by the light in the field of photonics.

• *Chapter 5* demonstrates that the magneto-elastic hybridization can expand the degree of freedom of spin waves by using mode degree of freedom of phonon.

When the light wave incidents to the boundary between different materials, the ray of light may split into two rays with orthogonal polarizations due to the optical anisotropy. In the case of reflection, there are few such phenomena that the ray of light splits. Magnon-phonon hybridized waves have a mode degree of freedom inherited from phonon, and the modes are mixed to each other at the sample edge because of the breaking of translational symmetry. We experimentally investigated the propagation and reflection dynamics of magnon-phonon hybridized waves in the vicinity of the sample edge. Interestingly, the hybridized waves split into two during reflection at the sample edge, exhibiting unconventional propagation dynamics. Spectroscopic analysis on the phenomena reveals that the split occurs owing to the mode degree of freedom of magnon-phonon hybridized waves is explained by phenomenological model considering elastic mode conversion and spin wave reflection. Our finding unveils the unknown role of magnon-phonon coupling in spin dynamics.

• *Chapter 6* shows the coherent interconversion between magnon and phonon owing to magnon-phonon coupling.

When two quasi-particles are coupled to each other, the interaction leads to the interexchange of the particles in time: Coherent oscillation. Owing to the lowest order of magnon-phonon coupling, magneto-elastic coupling, the magnon in a magnetic material may oscillates between magnon state and phonon state in time. This oscillation has yet to be observed since the magneto-elastic gap is in the range of several tens of MHz, which is much smaller than the frequency resolution of conventional spin-wave spectroscopy. In our approach, frequency resolution is defined by the length of measurement and thus could reach below 20 MHz, which allows the access to the magneto-elastic gap. We experimentally demonstrated that the magnetic garnet Lu₂Bi₁Fe_{3.6}Ga_{1.4}O₁₂ has a gap at the crossing between dispersion relation of phonon and magnon, and moreover, we observed temporal oscillation of the magnon amplitude. The oscillation is explained by a model based on the coherent state of hybridized state, leading to the estimation of magneto-elastic coupling constant.

Our

finding paves a way to spectroscopic study of magneto-elastic coupling with high frequency resolution.

• *Chapter 7* introduces the machine learning technique that could accelerate the experiment using time-resolved magneto-optical microscopy.

Time-resolved magneto-optical imaging is a versatile and powerful technique for observation of spin waves. On the other hand, the disadvantage of the method is the measurement time. The acquisition of a single video takes typically more than a week, making the experiment inefficient. We developed the machine learning technique that can interpolate the video frame based on the estimation of the vector field that describes pixel-to-pixel correspondence between different frames by using Convolutional Neural Network. The developed technique demonstrates a better interpolation quality compared with the previous state of art.

• Chapter 8 is devoted to summarize our results and comment on their importance.

The results obtained in this study are important steps towards a complete physical picture of the role of phonon to spin dynamics in magnetic insulators. The measurement technique established as a time-resolved magneto-optical imaging enables direct observation of the spatio-temporal spin dynamics, which revealed reflection and refraction law of magnon-phonon hybridized waves. Besides, the method is powerful for spin-wave spectroscopy with extremely high frequency resolution which has not reached by other spectroscopic technique, so that the method revealed the existence of magneto-elastic gap and coherent oscillation between magnon and phonon.

題目:時間分解磁気光学イメージングを用いたマグノン–フォノン混成波ダイナミクスの 研究

スピン波は磁性体中の磁化歳差運動の波動であり、磁性体の磁気応答を支配している。 スピン角運動量を利用したエレクトロニクスであるスピントロニクスでは、薄膜やナノ加 工された微小スケールの磁性体中のスピン波の生成とその制御が機能創成の鍵を握る。微 小磁性体に特徴的なスピン波は、膜厚などの磁性体の長さスケールと同程度の波長を有し ている。この波長領域では磁性体の形状に強く依存した静磁相互作用がスピン波の分散を 複雑にし、更にフォノンと混成することで多彩なダイナミクスが現れることが期待されて いるが、これらを実験的に直接観測することは難しかった。本論文では、薄膜磁性体中の スピン波の実時間実空間ダイナミクスを超高速レーザー分光法によって調べ、フォノン混 成効果や境界面効果を明らかにすることを目的としている。

本研究ではガーネット系磁性絶縁体薄膜に集光フェムト秒パルスレーザーを照射する ことでスピン波を励起している。この際焦点形状によりスピン波の伝播方向を任意に制御 することが可能であり、励起されたスピン波の伝播を時間分解磁気ファラデー顕微法によ り実空間像として取得することに成功した。時間空間像をフーリエ解析することにより、 スピン波とフォノンの分散の同時取得に成功した。その結果、スピン波のフォノン混成に よる長寿命化、さらにスピン波・フォノン混成状態におけるスピン波・フォノン間のコヒ ーレントな振動現象を見出した。これは、スピン波とフォノンがコヒーレントな相互作用 をしている決定的な証拠である。また、試料端面において通常の反射とは異なる波動ベク トルをもった反射スピン波が現れることを発見し、これが静磁相互作用の効いたスピン波 とフォノンの混成効果による複反射効果であることを突き止めた。更に、静磁相互作用と フォノン効果を取り入れたスピン波の数値計算を行い、実験結果を系統的に説明すること にも成功している。これにより、静磁相互作用やフォノンを利用してスピン波を精密に制 御する学問体系が構築された。

日置友智氏提出の論文は、薄膜磁性体中のスピン波の新しい分光学的研究法を開拓し、 スピントロニクスに不可欠な薄膜スピン波物性物理分野を切り拓くものであり、高く評価 できる。この成果は、提出者の日置友智氏が高度の学識と自立して研究する能力があるこ とを示すと判定される。よって、博士(理学)の学位論文として合格と認める。