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論　文　内　容　要　旨

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学位論文の題　　目	Magnetic Properties and Spin Excitations in Skyrmion Lattice Phase of the Itinerant Chiral Magnet $MnSi_{1-x}Ge_x$ (遍歴スカーミオン格子磁性体 $MnSi_{1-x}Ge_x$ の磁気特性とスピン励起)		

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Abstract

MnSi has a cubic chiral structure with the noncentrosymmetric space group $P2_13$. Due to the lack of inversion symmetry at the center of the Mn-Mn bond, the antisymmetric spin-spin interaction, called Dzyalonshinskii-Moriya (DM) interaction, leads to the long-period modulation to otherwise ferromagnetic structure. The resulting long-period helically modulated structure is established below T_c ~ 30 K under zero external magnetic field. Interestingly, the magnetic properties of MnSi can be tuned by the atomic substitution. It was reported that Fe, Co, and Ir-substitutions (positive chemical pressure with electron doping) on MnSi lead to the suppression of the ordering temperature T_c. In contrast, Al and Ga-substitutions (negative chemical pressure with hole doping) on MnSi lead to the increase of T_c. Notably, both types of chemical substitutions with electron/hole doping tends to increase the width of the skyrmion phase region. The enhancement of this skyrmion phase stability is not well understood for both types of substitutions. In contrast to the various elemental substitutions described above, the Ge-substitution to the Si site has prominent advantage; the Ge substitution only gives rise to the

negative chemical pressure, but no electron/hole doping as Ge is isovalent to Si. Hence, we studied the Ge-substitution effect to MnSi in detail, to elucidate the key parameters for the enhancement of the skyrmion phase stability.

MnSi also attracts renewed interest because of the discovery of the skyrmion-lattice structure under finite magnetic field. The magnetic skyrmion is a topological spin texture made of swirling magnetic moments. Recently, the spin excitations, so called `magnons', in such spin texture was studied theoretically and was found that topological nature of skyrmion will give non-trivial topological number (Chern number) for each magnon bands, resulting in the formation of the topological magnon bands. Such magnon band had been studied by means of inelastic neutron scattering one year after the skyrmion phase was found. However, the energy resolution was not sufficient to capture the magnon band splittings predicted by the theory. Hence, we will investigate such magnon bands in the skyrmion lattice of MnSi and Ge-doped MnSi by performing inelastic neutron scattering experiment with ultra-high energy resolution.

The objectives of this thesis are: (1) to investigate the effect of Ge-substitution on magnetic properties in $MnSi_{1-x}Ge_x$, and (2) to investigate the spin excitations in the skyrmion lattice phase of MnSi and $MnSi_{0.98}Ge_{0.02}$.

The polycrystalline alloys of $MnSi_{1-x}Ge_x$ with nominal Ge concentrations up to x_0 = 0.15 were prepared by Arc- and RF melting methods, followed by appropriate thermal treatments under Ar-atmosphere. Single crystal samples of MnSi and $MnSi_{0.98}Ge_{0.02}$ were prepared by the Bridgmann method. The samples were characterized using powder X-ray diffraction and scanning electron microscopy, and their magnetic properties were investigated by using the SQUID magnetometer. Neutron diffraction experiments were performed using the cold-neutron triple-axis spectrometer (CTAX) at HFIR, ORNL, whereas in elastic neutron scattering experiments were performed using the cold-neutron triple-axis spectrometer (SIKA) at OPAL, ANSTO.

In the experimental results, we found that the solubility limit of Ge under ambient pressure is x = 0.144(5) with annealing temperature T_{an} = 1073 K. Magnetization measurements on $MnSi_{1-x}Ge_x$ samples show that increases rapidly in the low-x range, whereas it becomes saturated at higher concentration $x > 0.1$. The saturation moment M_s and the width of the skyrmion phase also increases as Ge-concentration increases. We, furthermore, observed the nonlinear behavior of magnetic modulation vector k as a function of Ge concentration. In the inelastic neutron scattering experiment on MnSi and $MnSi_{0.98}Ge_{0.02}$, the low-energy magnetic excitation modes were observed at several Q-points. We found that the magnetic excitation spectra are considerably broader than the energy resolution (~ 40μeV). In addition, the inelastic spectra are more weaker and broadened at higher Q-positions. We confirmed that this excitation is intrinsic to the skyrmion-lattice phase, by comparing it to the excitation spectra both in the fully-polarized and helical phases. On the other hand, we found the inelastic spectra in $MnSi_{0.98}Ge_{0.02}$ are more broadened than those in MnSi.

In the discussion, we extracted various magnetic parameters including saturation moment M_s, ordering temperature T_c, and critical field to the fully polarized state H_{c2} from the bulk magnetic measurements of $MnSi_{1-x}Ge_x$. Then using the mean field approximation, we estimated the exchange interaction, Dzyaloshinskii-Moriya (DM) interaction, and magnetic modulation vector k at $T = 5$ K. We found the enhancement of estimated exchange and DM interaction by Ge-substitution indicating the enhancement of density of states and the spin-orbit coupling. In addition, the estimated magnetic modulation vector k at $T = 5$ K shows qualitatively consistent behavior with the one observed in the neutron diffraction study at $T = 30$ K except for the apparent shift due to the temperature difference. However, the increase of k at higher x is weaker in the mean field estimation than the neutron observation. This indicates that there should be an intrinsic enhancement of spin fluctuations, which are in principle ignored in the mean field approximation. It should be noted that for large x the width of skyrmion-phase region becomes larger. Since it is fluctuation effect beyond mean field level that stabilizes skyrmion lattice phase, the large spin fluctuation is also suggested from this observation. Furthermore, by Ge-substitution, we confirm that the chemical disorder also contributes to the skyrmion phase stability. The study of magnetic excitations in MnSi and $MnSi_{0.98}Ge_{0.02}$ shows the broadening of inelastic spectra for the low-energy magnetic excitation modes. From the spin dynamics simulation, we suggest that the thermal fluctuation and the overlapping between two or more magnon bands at low energy region lead to the observed broadened inelastic spectra. In addition, more broadened inelastic spectra in $MnSi_{0.98}Ge_{0.02}$ compared to MnSi suggested the enhancement of spin fluctuation. On the other hand, the decreased intensity at higher Q-positions may be due to decreasing magnetic structure factor, a reminiscent of the ferromagnetic spin wave dispersion, which behaves as $\omega \sim q^2$.

The conclusions of this thesis are the solubility limit of Ge in $MnSi_{1-x}Ge_x$ under ambient pressure synthesis is found as $x = 0.144(5)$ with the annealing temperature $T_{an} = 1073$ K. By Ge-substitution, we found the enhancement of density of states and the spin-orbit coupling, which control the critical temperature T_c and modulation vector k. On the other hand, the skyrmion phase stability is affected by the spin fluctuations and the chemical disorder. Furthermore, the obtained spin excitations spectra in the skyrmion lattice phase correspond to the thermally broadened topological magnon.

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論文審査の結果の要旨

題目：遍歴スカーミオン格子磁性体 $MnSi_{1-x}Ge_x$の磁気特性とスピン励起

Aji Seno 氏提出の論文は磁気スカーミオン格子相を発現するカイラル磁性体 MnSi の磁気特性に対する Ge 置換効果、およびスカーミオン格子相中の磁気ダイナミクスの解明を目指したものである。

スカーミオンは連続場中に形成されるトポロジカル欠陥の一種であり、近年カイラル磁性体において磁気モーメントの配列に同様な欠陥構造が見られることが発見され、世界的に精力的に研究が進められている。中でも、カイラル磁性体 MnSi は最初にスカーミオン相が観測された系であり継続的な興味が持たれている。MnSi の磁気スカーミオンは三角格子相として発現するが、このスカーミオン格子相は磁気転移温度近傍の非常に小さな温度・磁場範囲でのみ形成することが知られていた。本論文では MnSi の Si を Ge で部分置換することで系に化学的圧力を加え、磁気スカーミオン格子相の安定性増強を試みている。詳細な研究の結果、Ge 置換により磁気転移温度等は単調に増加する一方、ヘリカル相の変調ベクトルは非単調な振る舞いを示すなど興味深い結果が得られている。また、この Ge 置換に伴うハミルトニアンパラメータの変化がスカーミオン格子相の安定化に寄与することが実験的に確認された。

他方、スカーミオン格子相におけるスピンダイナミクスも興味深い研究対象である。本論文では MnSi 及び Ge 置換 MnSi1-xGex (x = 0.02) の高分解能中性子非弾性散乱を行いスカーミオン格子相における磁気非弾性散乱スペクトルを調べている。そこでは、分解能に比較してブロードな磁気励起スペクトルが観測された。散乱強度は高波数領域にいくにつれ減少している。古典モンテカルロ法とランダウ・リフシッツ運動方程式計算を組み合わせたスピンダイナミクスシミュレーションに基づく定性的な議論から、幅の広い磁気励起の起源が温度によりブロードになった協力的磁気励起であることが提案された。

これらの研究成果は MnSi 磁気特性に対する Ge 置換効果のメカニズム解明、並びにこの系で発現する磁気スカーミオン相におけるスピンダイナミクス理解に対する重要な指針となるものである。このことは、本人が自立して研究活動を行うために必要な高度な研究能力と学識を有していることを示している。したがって、Aji Seno 提出の論文は、博士（理学）の学位論文として合格と認める。