A comparison between Si substitution and pressure effects on structural and magnetic properties of tetragonal Mn_3Ge

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Mn-based materials with tetragonal structure have attracted much attention in the flied of spintronics and permanent magnet [1,2], because these alloys exhibit high magnetic transition temperature and strong uniaxial magnetocrystalline anisotropy without 4*f* elements. Among them, Mn_3Ga and Mn_3Ge with tetragonal $D0_{22}$ structure have crystallographically different two Mn sites, Mn_I and Mn_{II} sites. The magnetic moment at Mn_I site (Wycoff position 2*b* site) is directed opposite to that at Mn_{II} site (4*d* site), leading to a ferrimagnetic ordering with low magnetization of the alloys. It has been known that the magnetic properties of tetragonal Mn alloys are strongly influenced by the number of the valence electron [3,4]. On the other hand, theoretical investigations have predicted that the magnetic state corresponding to exchange interactions between the Mn sites is sensitive to structural properties such as an atomic order, an off-stoichiometry and an atomic distance [5,6]. In this study, we have investigated Si substitution and pressure effects on magnetic properties of off-stoichiometric Mn_3Ge with tetragonal $D0_{22}$ structure to clarify relationship between magnetic and structural properties of the alloys.

Polycrystalline samples of $Mn_{3.09}Ge_{0.91-x}Si_x$ were prepared by arc-melting method. Powder samples prepared from the ingots were aged at 673 K for a week to obtain the tetragonal $D0_{22}$ phase. Structural and magnetic properties at ambient pressure were investigated by a conventional powder X-ray diffractometer and a vibrating sample magnetometer. Magnetization measurements under high pressure were performed using a superconducting quantum interference device and a piston-cylinder-type pressure cell with a liquid pressure-transmitting medium (Daphne 7373). Powder X-ray diffraction experiments at room temperature under high pressure were carried out using a diamond anvil cell with a liquid pressure-transmitting medium (methanol:ethanol = 4:1) and synchrotron radiation at Photon Factory BL18C in High Energy Accelerator Research Organization (KEK).

Powder X-ray diffraction data of $Mn_{3.09}Ge_{0.91-x}Si_x$ at ambient pressure show that a single phase of the $D0_{22}$ structure is obtained up to $x = 0.4$. The lattice constants *a* and *c* estimated from the X-ray diffraction data linearly decreases with increasing Si content. This is due to a substitution with Si atom having smaller ionic radius than Ge atom. As the result, the unit cell volume shrinks with the increasing Si content. The axial ratio *c*/*a* is mostly constant, indicating that the unit cell volume is isotropically contracted by the Si substitution. Results of high pressure X-ray diffraction experiments at room temperature indicate that the $D0_{22}$ structure of $Mn_{3.09}Ge_{0.91}$ is kept up to 10 GPa. The lattice constants *a* and *c* and the unit cell volume monotonically decrease with increasing pressure. However, the axial ratio *c*/*a* is not affected by applying pressure. These results indicate that the pressure effect on the structural properties of $D0_{22}$ structure is mostly same to the Si substitution effect. Magnetization of $Mn_{3.09}Ge_{0.91-x}Si_x$ at ambient pressure decreases with increasing Si content. Similarly, magnetization of $Mn_{3.09}Ge_{0.91}$ decreases with increasing pressure, as shown in Fig. 1. The unit cell volume dependence of magnetization for $Mn_{3.09}Ge_{0.91-x}Si_x$ is consistent with that for $Mn_{3.09}Ge_{0.91}$ under high pressure. From the results of the Si substitution and high pressure experiments, we can conclude that the Si substitution and pressure effects on magnetization are attributed to the structural properties of $D0_{22}$ structure such as contraction of unit cell volume and shrinkage of atomic distance between Mn atoms.

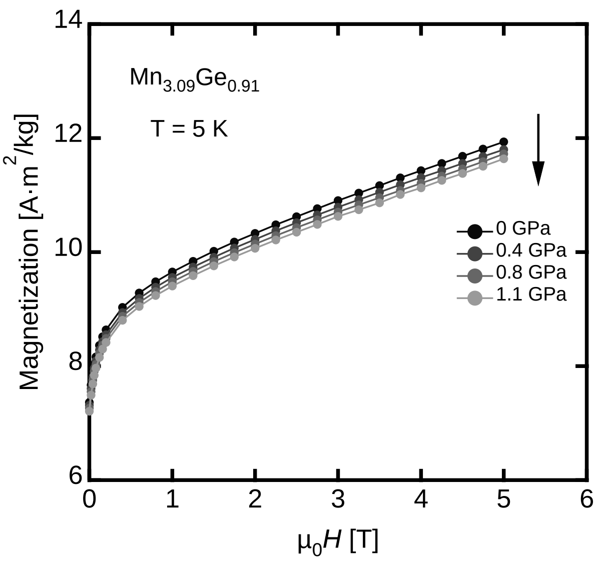

Figure 1. Magnetization curves at 5 K under various pressures up to 1.1 GPa for $Mn_{3.09}Ge_{0.91}$.

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