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Author	寺田, 道子(Terada, Michiko) 坂田, 亮(Sakata, Makoto)
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Ferromagnetic Resonance in Thin Films of Mn_5Ge_3 at *X*-Band

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Michiko TERADA*
Makoto SAKATA**

Abstract

Polycrystal thin films of intermetallic compound Mn_5Ge_3 are prepared by the flash evaporation technique and their magnetic properties are investigated through the ferromagnetic resonance at *X*-band (9380 MHz) at various temperatures. The resonance conditions derived by Kittel are applied. The effective magnetization is smaller than that of the bulk material. *g*-factor is constant of 2.1 between 15°C and 45°C, but increases sharply near 0°C. The observed line width increases with decreasing temperature. Below 0°C, the abnormal resonance was observed. These phenomena are considered to be due to the increase of crystal magnetic anisotropy of Mn_5Ge_3.

I. Introduction

The existence of ferromagnetic resonance (FMR) was predicted by Landau and Lifshitz on ferromagnetic permeabilities in 1935.[1] The theory was extended by Kittel[2),3)] and he showed the resonance conditions which depend on the demagnetizing factor and, in a single crystal, the crystalline anisotropy. Further, he proposed that it is possible to excite the spin wave resonance by a *rf* field in a ferromagnet of single crystal.[4)]

A number of investigations of FMR have been made for metals and alloys,[5),6)] but few of them for intermetallic compounds. The Mn_5Ge_3 is an intermetallic compound with the crystal structure of $D8_8$ type,[7)] which is illustrated in Fig. 1, forms a solid solution within a few percent of manganese[8)] which is called "η-phase" by Ohoyama,[9)] and is a ferromagnet with the Curie temperature of 27°C for the bulk material.[10)] The observation of FMR was made at room themperature.[11)] The experimental results showed that both the resonance fields and the half widths for various films were independent of the substrate temperature and the thickness of

* 寺田道子 Assistant, Faculty of Engineering, Keio University.
** 坂田亮 Professor, Faculty of Engineering, Keio University.

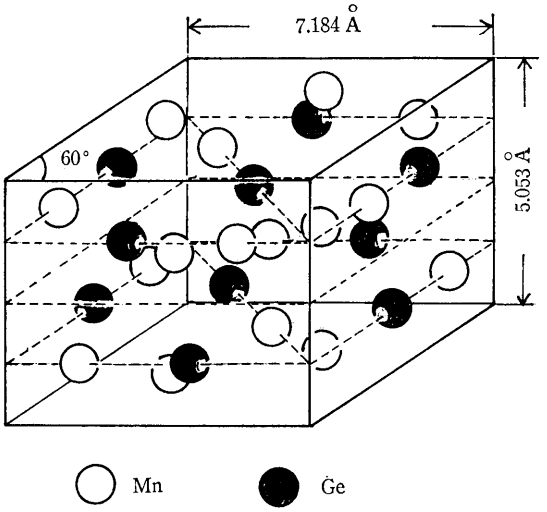


Fig. 1 Crystal structure of Mn_5Ge_3; $D8_8$ type.

films, and that the resonance field was dependent on the temperature. The estimated g-factor tends to increase slightly at lower temperatures. In this paper, the means of preparation of the films of Mn_5Ge_3 and the temperature dependence of FMR at X-band are reported and the magnetization, the g-factor and the line shape are discussed qualitatively.

II. Experimental Procedure

The thin films were prepared by the flash evaporation technique[12] in order to avoid the deviation from the stoichiometric composition. The particles of Mn_5Ge_3 used as the evaporant were fallen bit by bit to a heated filament by vibrating a sample hopper and evaporated in a moment on substrates. A conical basket of tungsten was used as a filament. The vaccum during evaporation was kept between 10^{-7} and 10^{-6} Torr by a sublimation pump. The substrates were cover glass plates (8 mm×8 mm) cleaned by a chemical treatment and rock salts with a cleavage surface. The substrate temperature was ranged from room temperature to 325°C. The thickness was measured by means of a multiple beam interferometer.

The crystal structure of films was examined through a transmission method of electron diffraction. The films thinner than 1000 Å were deposited on rock salts and stripped from the substrates. For the use of magnetic measurements, the films with the thickness from 1000 Å to 4700 Å were prepared on the cover glasses.

The investigation of FMR was made by means of a ferromagnetic resonance spectrometer using a TE_{102} cavity at 9380 MHz. The measurement temperature was

varied from liquid nitrogen temperature to 50°C. If the magnetic anisotropy is negligible, which is possible in the case of polycrystal substances, the resonance condition for a thin film is given by[2)]

$$\frac{\omega}{\gamma} = H_{\perp} - 4\pi M_s$$

with the static magnetic field perpendicular to the plane of film ($H_{\perp}$); and by

$$\frac{\omega}{\gamma} = \sqrt{H_{//}(H_{//}+4\pi M_s)}$$

with the static magnetic field parallel to the plane of film ($H_{//}$), with

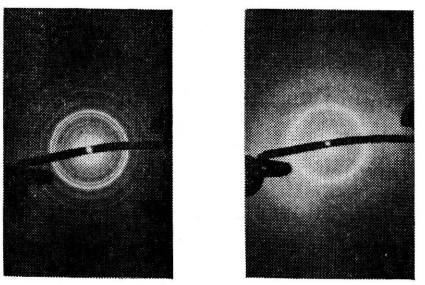
$$\gamma = \frac{ge}{2mc}$$

where ω is the angular frequency of rf field, M_s the saturation magnetization, g the g-factor and e, m, c are charge, mass, velocity of electron, respectively.

For the reference, the magnetization of bulk material in polycrystal was measured by means of a magnetic balance at temperatures between −190°C aud 45°C and at various magnetic fields.

III. Experimental Results

The electron diffraction patterns are shown in Fig. 2. It is found that the prepared films are polycrystal of η-phase with the structure of $D8_8$ type when the substrate temperature T_s is above 150°C (Fig. 2a). But the diffraction intensity suggests that the composition deviates slightly from the stoichiometric ratio. When the T_s is below 150°C, the diffraction lines are so haloed that it is impossible to know the crystal structure (Fig. 2b). It is supposed that the film for the T_s below 150°C consists of very fine grain size.



(a) (b)

Fig. 2. Electron diffraction patterns; (a) at T_s=320°C and (b) at T_s=140°C.

The conditions for the successful preparation of the Mn_5Ge_3 films were to use as the evaporant the particles between 40 and 300 mesh, to keep the filament temperature at about 2000°C, which is far higher than the melting point of 932°C, and to heat the substrate to temperatures above 150°C.

The FMR was observed on the films for the T_s above 150°C but was not below 150°C. This fact suggests that the films for the T_s below 150°C are non-ferromagnetic material because of fine grain size. The spin wave resonance was not observed on any films. Further, the observation of FMR was made at temperatures above 0°C since there appeared abnormal phenomena at lower temperatures than 0°C; that is, the half width of line shape was broaden and that the line shape deviated from the normal shape with the variation of temperature as shown in Fig. 3. It seems that the line shape of differential curve is transformed into the absorption curve at about −20°C when the static magnetic field was perpendicular to the plane of the film and that the spectrum intensity disappears at nearly −22°C when the magnetic field was parallel to the plane of the film. Furthermore, an anomalous peak was observed at temperatures below 12°C near the resonance field.

The experimental results above 0°C are as follows. The temperature dependence of the resonance field and the half width are shown in Fig. 4 and Fig. 5. The value of $H_\perp$ decreases from 7000 Oe to 3500 Oe and $H_{//}$ increases from 500 Oe to 3000 Oe with increasing temperature from 0°C to 45°C. In Fig. 4, the resonance fields which are estimated from the saturation magnetization of bulk material assuming g-factor to be 2 are shown by a broken line.

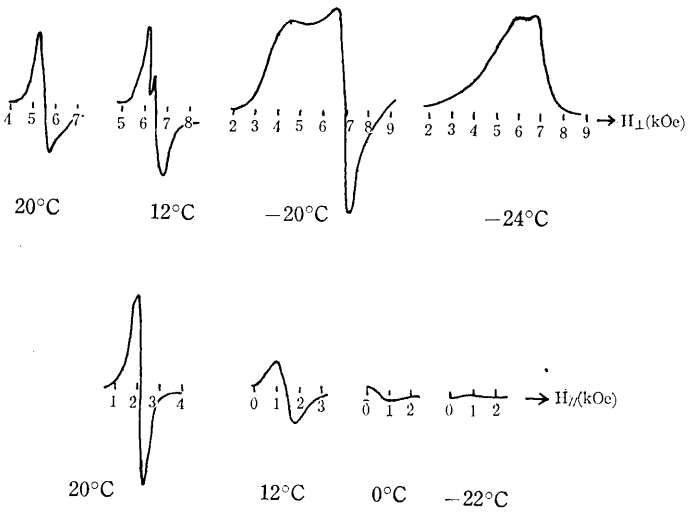


Fig. 3. FMR spectra at several temperatures of the film with thickness of 2600 Å and T_s=320°C.

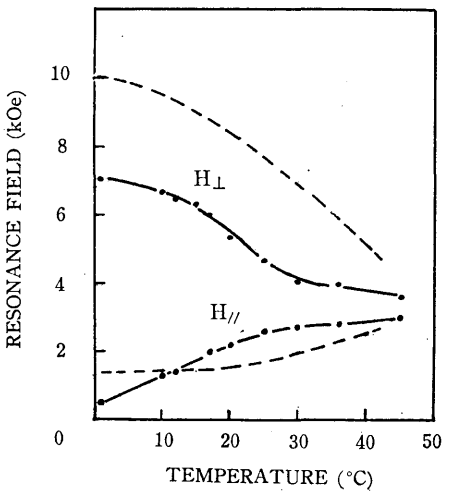


Fig. 4. Temperature dependence of resonance field. Broken line shows the resonance field estimated from the magnetization of bulk material at 9.4 kOe assuming g-factor to be 2.

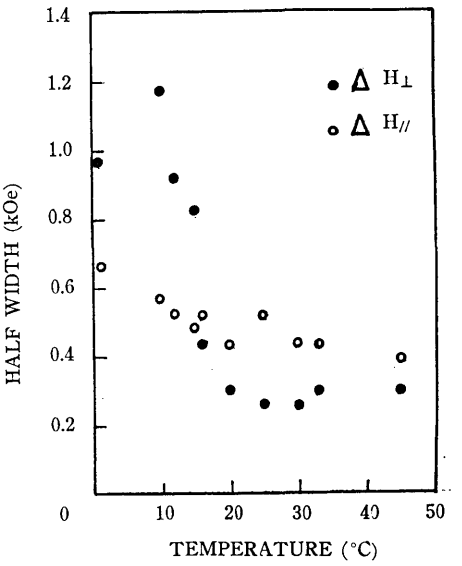


Fig. 5. Temperature dependence of half widths.

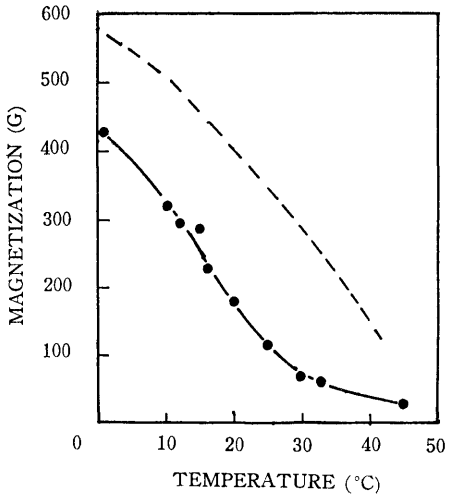


Fig 6. Magnetization estimated from the resonance conditions at various temperatures. Broken line shows the magnetization of bulk material at 9.4 kOe.

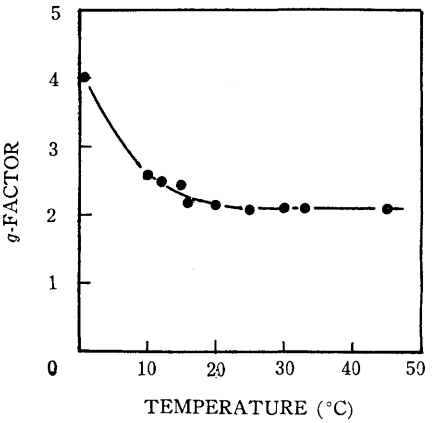


Fig. 7. g-factors estimated from the resonance conditions at vatious temperatures.

The half widths, $\Delta H_{\perp}$ and $\Delta H_{//}$, have a tendency to increase with decreasing temperature. The values of M_s and g-factor estimated from the resonance conditions are shown in Fig. 6 and Fig. 7 against temperature. In Fig. 6, the magnetization of bulk material at 9400 Oe are shown by a broken line for reference. The g-factor is constant about 2.1 between 15°C and 45°C, but increases sharply to reach about 4 at 0°C, which is extraordinarily large value.

IV. Discussion

The value of M_s are smaller than the magnetization of the bulk material at 9400 Oe. The critical temperature from the extraporation of the curve of M_s^2 vs. temperature is 22°C, which is slightly smaller than the Curie point of the bulk material. When the saturation magnetostriction constant (λ_s) and the planner stress in the film plane (σ) are taken into consideration to the resonance conditions, we can substitute the effective magnetization

$$M_{\text{eff}}=M_s+\frac{3\lambda_s\sigma}{4\pi M_s}$$

for M_s in the case of thin film. Therefore, the estimated magnetization of thin film may be smaller than the saturation magnetization if λ_s is negative. However, λ_s of Mn_3Ge_3 is unknown.

On the other hand, it is hard to explain such a large g-factor at 0°C of ferromagnetic materials. It is well known that the effective g-factor of the ferrimagnetic materials with the compensation point increases sharply near that point. The compound Mn_5Ge_3, however, has no a compensation point.

The line widths increased near 0°C. The causes of the line broadening were discussed by Van Vleck[13], Bloembergen[14] and others. The following are generally regarded as the mechanism of line width; (a) dipolar interaction, (b) spin-lattice interaction, (c) skin effect, (d) crystal anisotropy, (e) interaction of ferromagnetic spins with conduction electrons. However, except (d) these are considered to give the microscopic contribution to line broadening, at most, several Oersted in ferromagnetic materials. The crystal anisotropy, which was negrected in the applied resonance conditions, can give rise to the shift in the resonance field by an anisotropy field in single crystals, for example $2K/M_s$ in the uniaxial crystals with the anisotropy constant K. In polycrystals, it may contribute to the line broadening since the anisotropy axis may partly be in the different direction in the specimen. The line width due to the anisotropy is considered to be in the order of $2K/M_s$ in the case of the random orientation of anisotropy axis. In practice, it is hard to deduce the line width theoretically, as the degree of crystalization of present specimen is unknown.

An anisotropy constant, in general, increases with the rise of saturation magnetization in decreasing temperature. The temperature dependence of anisotropy of Mn_5Ge_3 in single crystal has been observed by Tawara et al.[15] They found that the direction of easy magnetization lies along the c-axis and that the anisotropy constant varies from 3.0×10^5 to 4.2×10^6 erg/cm^3 with decreasing temperature from room temperature to liquid nitrogen temperature. It is expected that such a strong anisotropy will give rise to line broadening unless the static magnetic field is high enough to saturate the magnetic moment. As the magnetization of the film was not observably by means of the present magnetic balance, that of polycrystal bulk material was measured for reference at various temperature and magnetic field, which is shown in Fig. 8 and Fig. 9. They show that the magnetic moment at −190°C is nearly saturated at 5400 Oe but that at 0°C it is not saturated at 7000 Oe. Thus the static magnetic field applied in the FMR might be not so high enough that the magnetization vector turns to the direction of the static magnetic field. It is supposed that the g-factors at temperatures just below the Curie point are estimated apparently large because the resonance conditions are applied to the unsaturated magnetization.

The deformed line shape and the anomalous peak may also be attributed to this strong anisotropy. These phenomena are analogous to the FMR in nickel single crystal at X-band at low temperature.[16] It is reported in the reference (16) that a full resonance curve could not be observed below −50°C, at about −160°C the

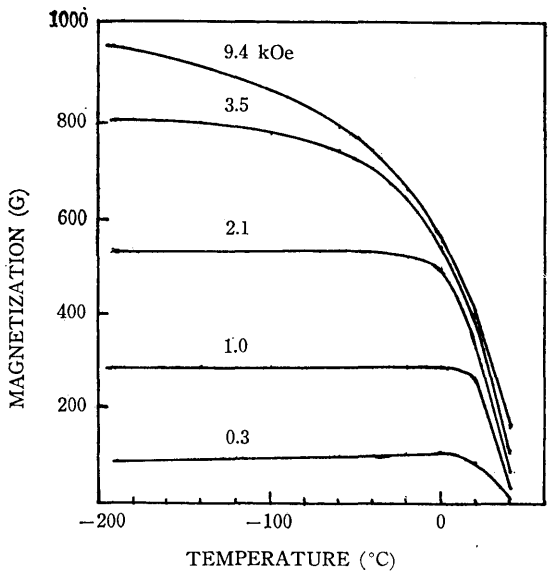


Fig. 8. Temperature dependence of magnetization for bulk material.

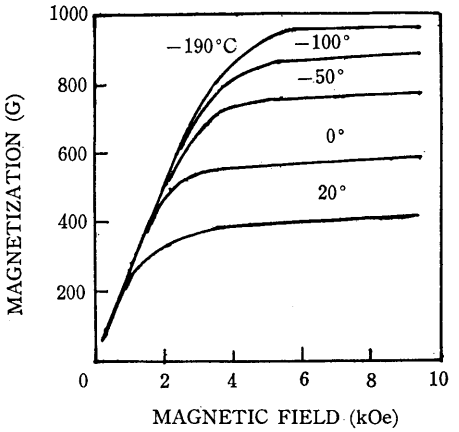


Fig. 9. Magnetization curves at several teperatures for bulk material.

resonance peaks had disappeared completely with the static magnetic field parallel to the easy direction of magnetization and that the secondary peak were observed. These facts are explained by the increasing of crystalline anisotropy. The possibility of the two resonances in the single crystal thin film at low frequency has been studied by several investigators[17),18)] through the single- or multi-domain structure, in which the direction of the magnetization vector does not coincide with that of the applied field because of an anisotropy field. As the present Mn_5Ge_3 films, however, are not single crystal, it is difficult to explain an anomalous peak by the simple single- or multi-domain structure.

The magnetization vector is probably not in the direction of the static magnetic field since the anisotropy of Mn_5Ge_3 increases sharply at temperatures lower than the Curie point. In order to saturate the magnetic moment, the observation of FMR has to be made at a higher frequency so that the resonance field becomes higher. The FMR measurement at Q-band ($f=34.7$ GHz) is in progress.

Acknowledgments

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