Frequency Dependence of FMR in Mn-Fe Ferrite Spheres

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Abstract

Resonance experiments of conductive manganese ferrites have been made at 14.8 GHz by using spherical samples which were examined previously at 9.3 GHz. The linewidth increases monotonically with temperature and does not exhibit any maxima between 77–300 K. This is quite different from the results at 9.3 GHz. The characteristic linewidth behavior at 14.8 GHz is ascribed to the excitation of surface modes near the bottom of the spin-wave manifold. The observed field for the main resonance maximum is well explained by considering the excitation of surface modes near the top of the manifold.

1. Introduction

Ferromagnetic relaxation has been widely investigated from both theoretical and practical points of view. A variety of relaxation mechanisms, each giving a finite FMR linewidth, have been thus proposed in a number of ferromagnets, for example, as seen in ref. (1) by Sparks. In non-stoichiometric ferrites containing excess iron, hopping motions of electrons between adjacent Fe^{2+} and Fe^{3+} ions are coupled to the precessional motion of magnetization so as to cause a typical slow relaxation.[2] Since the electron hopping motions, which occur as far as temperatures are not too low, should also cause electric conduction,[3] these ferrites can no longer be treated as insulators like stoichiometric ferrites. A care must be taken of the skin effect in making FMR measurements with them. This includes to elucidate its contribution carefully if the effect is inevitable.

In fact, the skin depth is fairly small in some conductive ferrites, in particular near room temperature. In such cases it is not easy to prepare spherical samples smaller than or even comparable with the skin depth. Typical examples are excess-iron manganese ferrites. The $Mn_xFe_{3-x}O_4 (x<1)$ system has been carefully studied at 9.3 GHz with samples of spherical,[4] cylindrical[5] and plate-like[6] shapes. It has been shown for spherical samples that the skin effect contribution yields even a linewidth maximum at low temperatures between 100–200 K, and that the temperature dependence of the resonance field near room temperature can well be inter-

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preted in terms of surface modes excited near the top of the spin-wave manifold. In view of these results, it is interesting to examine resonance behaviors at higher frequencies.

Recently Maryško[7] has performed FMR measurements on the same system at 7.8, 15.2 and 16 GHz. He has pointed out that the linewidth behaviors found at 15.2 and 16 GHz, which are somewhat different from those at 7.8 GHz, can be qualitatively understood as a result of the excitation of certain volume modes on the high field side of the main absorption maximum, and further that the appearance of an additional absorption maximum on the high field side can be interpreted also along the same line of argument.

This paper concerns similar FMR measurements, which were carried out at 14.8 GHz by using the same samples of manganese ferrite as used previously at 9.3 GHz.[4] As shown in a later section, we propose that the additional maximum should be connected with the surface modes which are excited at the bottom of the spin-wave manifold rather than with a magnetostatic volume mode as proposed by Maryško. Description will be made of the obtained results with emphasis for a dominant role of these surface modes on the linewidth behaviors.

2. Experimental

Spherical samples used in the present experiments were prepared from single crystals $Mn_xFe_{3-x}O_4$ grown by Verneuil's method. Three kinds of specimens were used in the measurements. The temperature dependences of conductivity (σ) of these specimens are well described by the formula[3]:

$$\sigma = (ne^2a^2/\tau_0 kT) \exp(-E_g/kT), \tag{1}$$

indicating that the conduction is caused by the electron hopping process. Here, n and a are the concentration and the jump distance of the hopping carriers, respectively. The activation energy E_g of specimen is shown in Table I, together with the value of x estimated from the resistivity ρ by using $a=3$ Å and $\tau_0=3\times10^{-13}$ sec.[3] in eq. (1). For specimen (1), the temperature dependences of $4\pi M_s$, K_1/M_s and ρ are also shown in Table II. Resonance experiments were carried out with ordinary spectrometers at 9.3 and 14.8 GHz between 77–350 K. In all cases, dc magnetic field was applied in a [111] direction.

Table I Resistivity and composition of the specimens.

Specimen	Resistivity at 300 K (Ω cm)	Activation energy (meV)	Composition x
1	0.28	40	0.95
2	0.12	30	0.91
3	0.08	40	0.78

Table II Temperature dependence of $4\pi M_s$, K_1/M_s and ρ for the specimen (1).

Temperature (K)	$4\pi M_s$ (G)	K_1/M_s (Oe)	ρ (Ωcm)
300	5200	−46	0.28
200	6300	−130	0.42
100	7000	−200	2.00

3. Experimental Results and Discussion

In Figs. 1, 2 and 3 are shown the temperature dependences of the resonance field (H_{res}), the linewidth (ΔH) and the lineshape at frequencies 9.3 and 14.8 GHz. The results at 9.3 GHz are the same as those reported previously[4] and are shown here for comparison with the results at 14.8 GHz. The distinct features at 14.8 GHz are as follows: (1) The size dependences of both H_{res} and ΔH are much smaller as compared with those at 9.3 GHz. (2) ΔH increases monotonically with temperature and does not exhibit any maxima between 77–300 K. (3) The asymmetry of lineshape at 14.8 GHz is quite different from that at 9.3 GHz. These behaviors are similar to those at 15.2 GHz reported by Maryško.[7] The resonance fields at 14.8 GHz well agree, over the whole temperature range, with those calculated by the resonance condition[4]:

$$(\omega/\gamma)^2=H_i(H_i+4\pi M_s)\,, \tag{2}$$

$$H_i=H-4\pi M_s/3-1.33\ K_1/M_s \tag{3}$$

for surface modes at the top of the spin-wave manifold (solid curve in Fig. 1). At 9.3 GHz, on the other hand, the observed resonance fields agree with eq. (2) only in the high temperature region.

For both frequencies the absorption curve at higher temperatures has a terraced shape on the high field side of resonance peak (Fig. 3). Clearly, the edge of the terrace yields a separate peak if the absorption is recorded by its derivative as is often done (Fig. 4). It is to be noted here that at 14.8 GHz the half-power point of absorption on the high field side falls on this terraced portion. In other words, the value of linewidth at this frequency depends predominantly upon the separation of the terraced portion from the field for the main absorption maximum. In our previous note[5] it was suggested that the edge of terrace, indicated as H_2 in Fig. 4, could be considered to correspond to a resonance field of the surface modes excited, for a very small skin depth, at the bottom of the spin-wave manifold, i.e., for the modes traveling along the dc magnetic field. For spheres the dispersion relation of the spin-wave is of the form

$$\omega_s(\theta)=\{(H-4\pi M_s/3)(H-4\pi M_s/3+4\pi M_s\sin^2\theta)\}^{1/2}, \tag{4}$$

if the exchange effect is omitted ($k\fallingdotseq 0$), where θ is the angle between dc magnetic

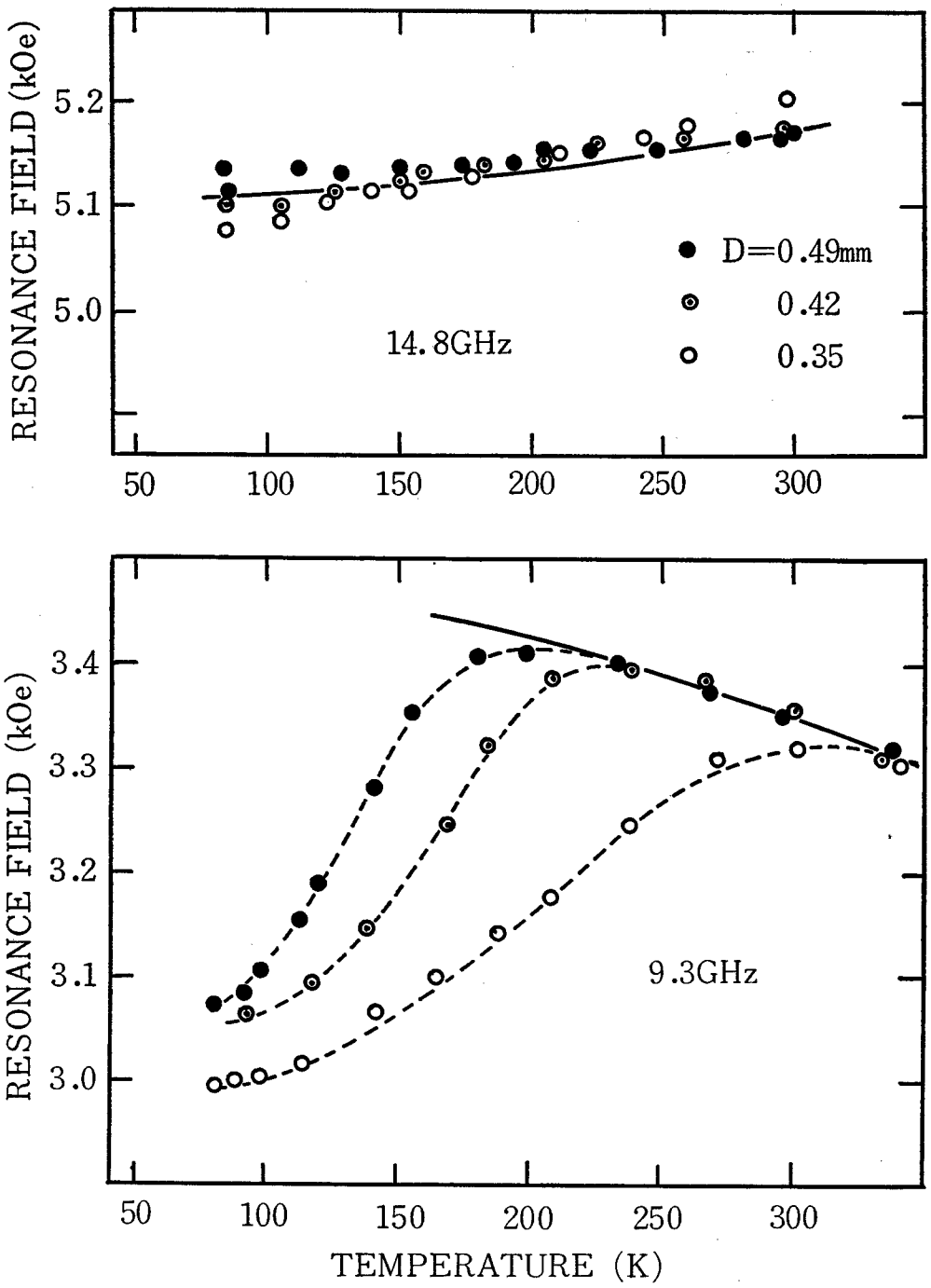

Fig. 1. Temperature dependence of the resonance field in a [111] direction for the specimen (1). Solid curves are calculated from eq. (2) for the cases of $g=1.92$ (14.8 GHz) and 1.94 (9.3 GHz).

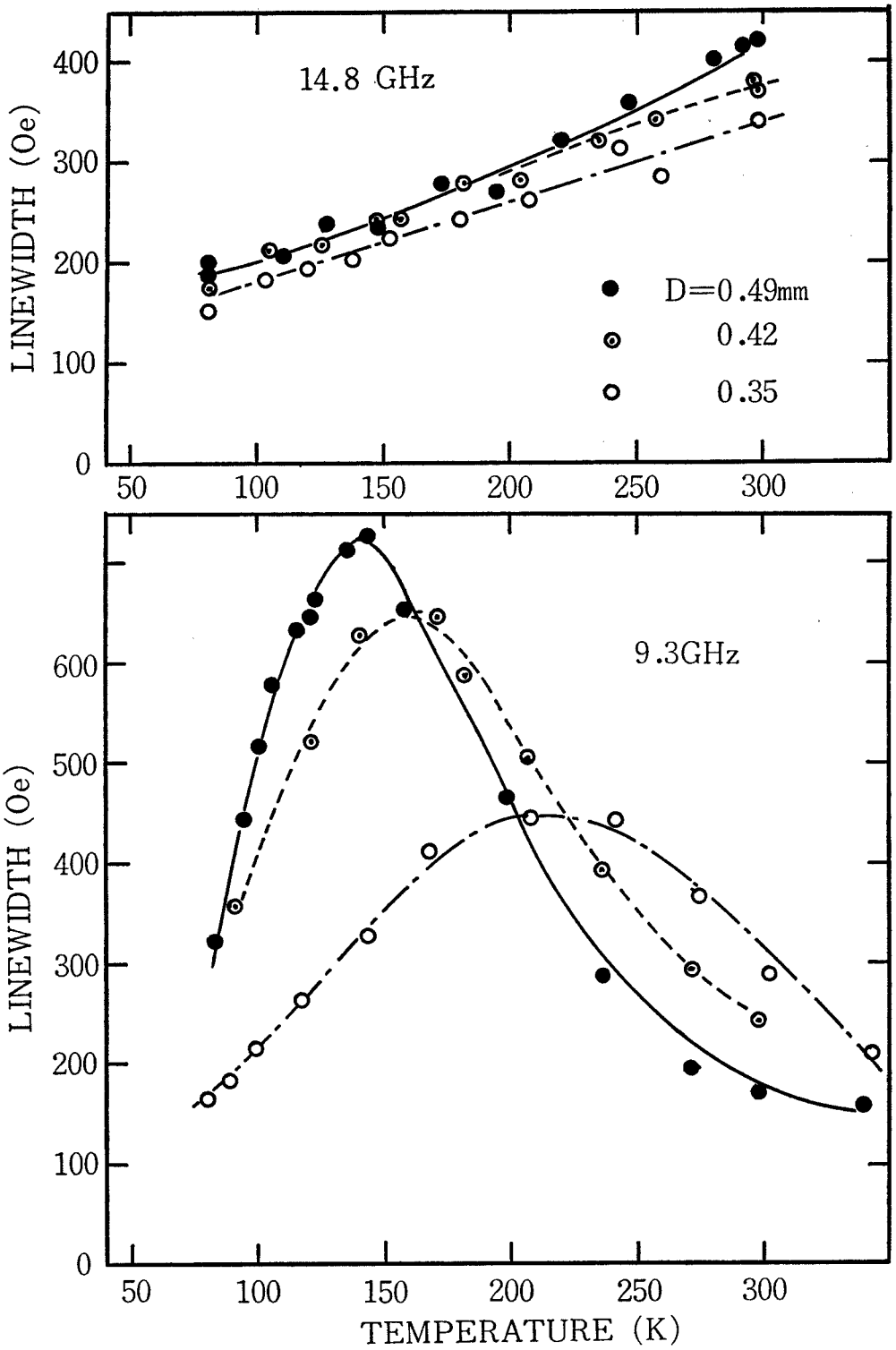

Fig. 2. Temperature dependence of the linewidth for the specimen (1).

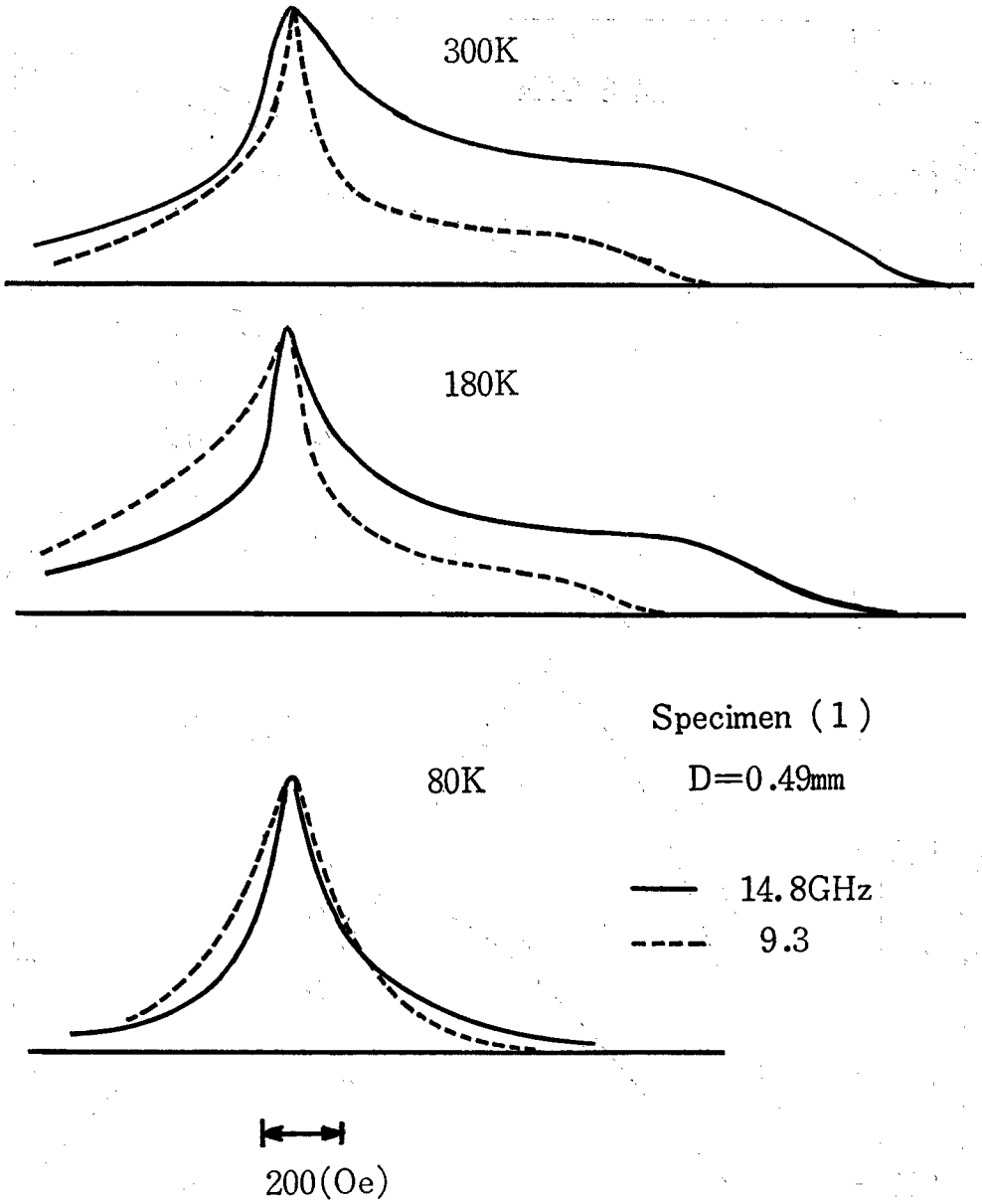

Fig. 3. Variation of lineshape with temperature at 14.8 and 9.3 GHz.

field and wave vector **k**. Here, the field (H_0) for the main absorption maximum is determined by $\omega_s(90°)=\omega$. While, the resonance field H_{s0} of the surface modes at the bottom of the spin-wave manifold is determined by $\omega_s(0°)=\omega$. The separation $\Delta=H_{s0}-H_0$ is then written, for a small skin depth, as

$$\Delta=(\omega/\gamma+2\pi M_s)-\sqrt{(\omega/\gamma)^2+(2\pi M_s)^2}\,. \tag{5}$$

Figure 5 shows the observed field separation between terrace edge and main maximum

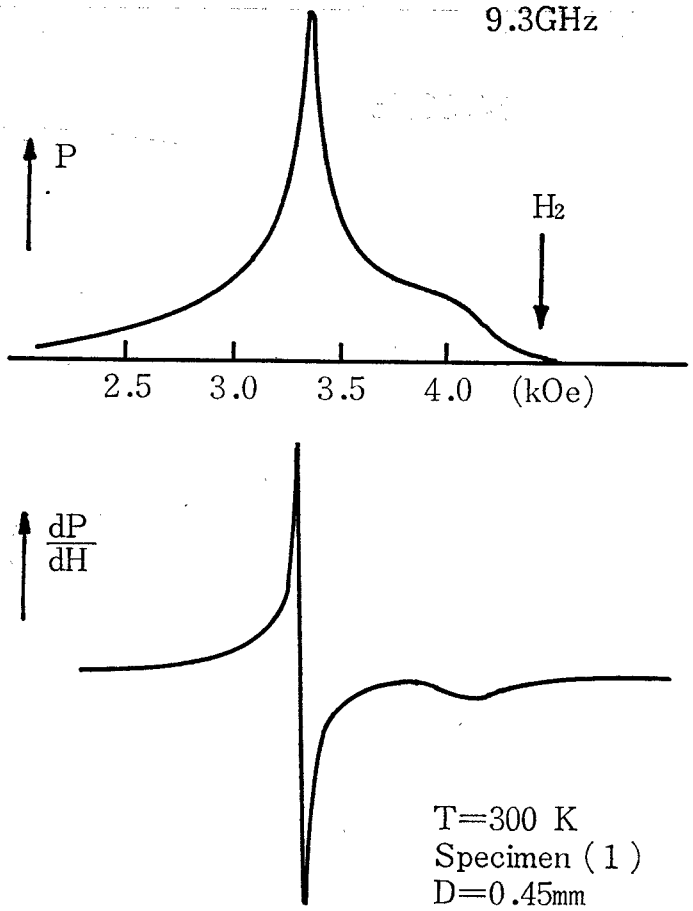

Fig. 4. Absorption curve at 9.3 GHz and its derivative.

as a function of D/δ, normalized by the value of Δ evaluated from eq. (5), where D is the diameter of sphere and δ is the so-called skin depth given by $\delta=c/\sqrt{2\pi\omega\sigma}$. For a sufficiently large value of D/δ, the position of the terrace edge well agrees with the bottom of the spin-wave manifold. With an intermediate value of D/δ the observed value of H_2-H_0 exhibits an appreciable deviation from the calculated value of Δ, presumably because the approximation of the small skin depth is no longer valid. However, we thus find that the edge of absorption around H_2 is essentially a characteristic feature of the surface phenomenon which may appear most typically in the case of a small skin depth. Maryško ascribed this anomaly around H_2 to the excitation of a magnetostatic volume mode.

On the other hand, the linewidth measured for various values of D/δ are shown in Fig. 6. The value of ΔH tends to increase as resonance approaches a surface type one. As seen in Figs. 5 and 6, this increase arises mainly from the increase in H_2-H_0 with D/δ. A terrace of absorption curve on the high field side appeared also at 9.3 GHz. But, in this case, the terrace was less prominent and had no ap-

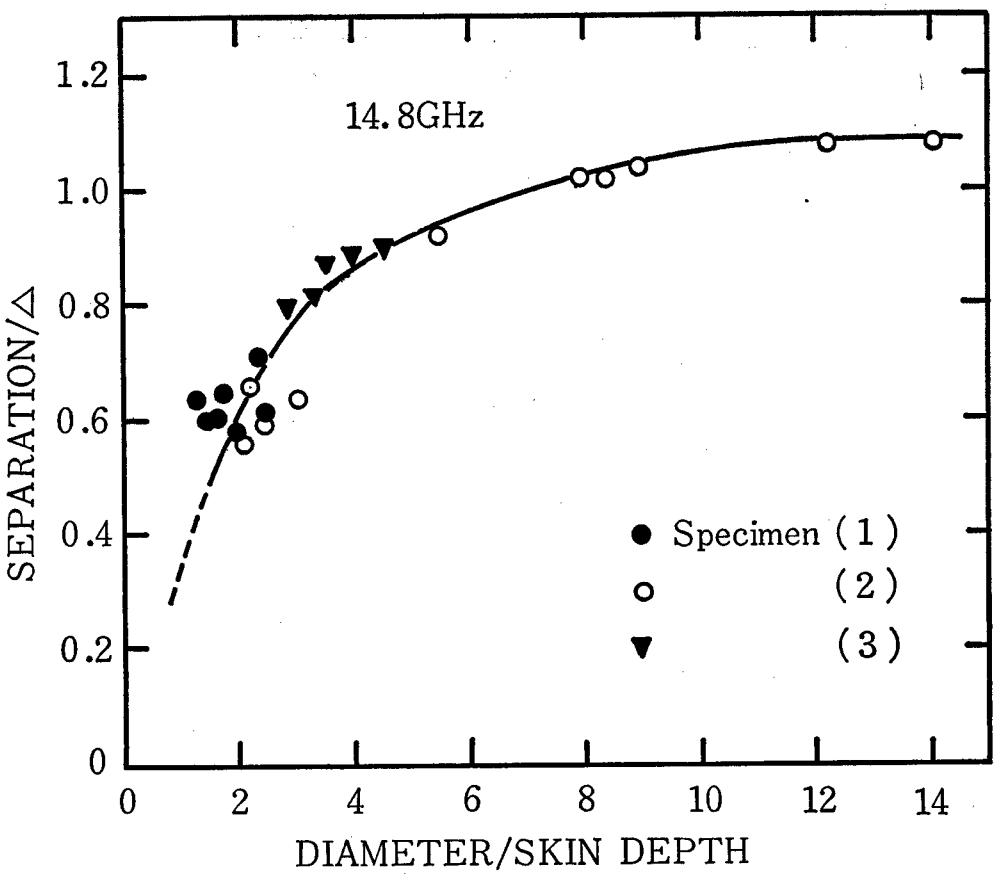

Fig. 5. Separation of terrace edge measured from the main absorption maximum. Δ is evaluated from eq. (5).

preciable influence upon the linewidth. As seen in Fig. 3, the absorption of the high field side at 14.8 GHz is much enhanced in comparison with that at 9.3 GHz. Maryško considered that the large absorption of the high field side would be connected with the excitation of some volume modes. On the basis of such a consideration it should be expected that the increase of sphere diameter suppresses the excitation of the volume modes and thus reduces the absorption of the high field side. In our experiments, however, the linewidth increased monotonically with the diameter. This means that the absorption between H_0 and H_{s0} is not reduced as expected above. Of course, it seems very likely, as pointed out by Maryško, that the asymmetry of lineshape depends crucially upon the location of the resonance field of the uniform mode (H_u) relative to that of the surface mode at the top of the spin-wave manifold (H_0), i.e. upon the sign of $H_u - H_0$. This idea was supported also in our experiments, for example, by a fact that the asymmetry at 9.3 GHz was reversed in the high temperature region where the sign of $H_u - H_0$ changes owing to the decrease of $4\pi M_s$. However, any detailed explanation of the large absorption

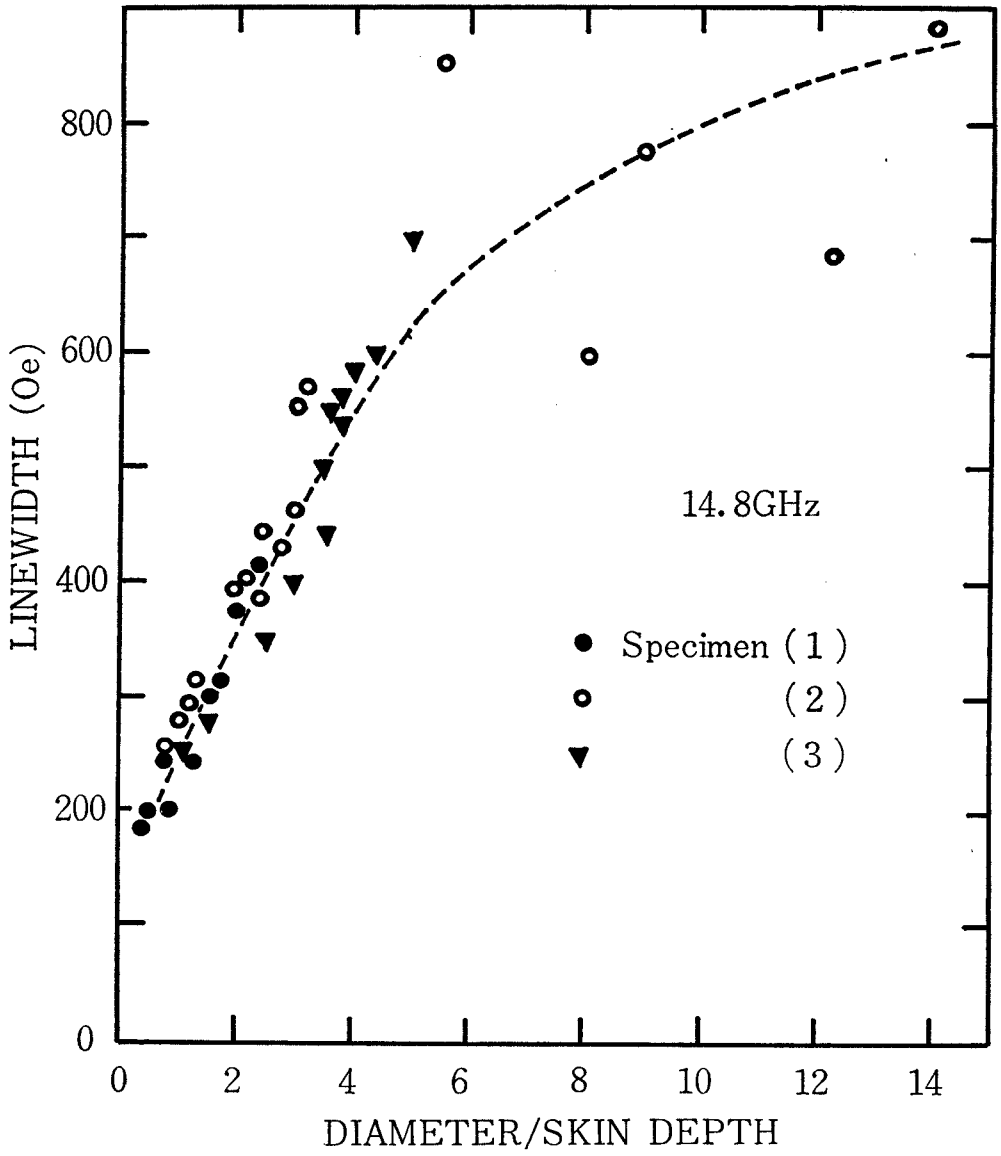

Fig. 6. Variation of linewidth with the ratio of sphere diameter to skin depth (D/δ).

on the high field side of the main maximum at 15 GHz has not yet been obtained.

Finally our conclusions are summarized as follows: (1) The edge of the terraced absorption around H_2 corresponds to the excitation of surface modes near the bottom of the spin-wave manifold. (2) The characteristic temperature dependence of linewidth at 15 GHz originates mainly from the D/δ—dependence of the resonance field for these surface modes, which was not of primary importance at 9.3 GHz in understanding the observed $\Delta H-T$ curves. (3) The main resonance maximum can be well understood by considering the excitation of surface modes near the top

of the manifold.

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