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SOME FEATURES OF MULTIPLET FERROMAGNETIC RESONANCE IN FERRITES

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Preliminary results are presented of the observation of multiplet ferromagnetic resonance in inhomogeneously magnetized single-crystal and polycrystalline ferrite samples. The inhomogeneous magnetizations required for the observation of this effect were produced mainly by the inhomogeneity of the demagnetizing fields of the investigated samples. Some features are noted of the absorption spectra observed under various conditions.

1. INTRODUCTION

USUALLY in the interpretation of experimental results on ferromagnetic resonance in magnetic semiconductors it is assumed that the magnetic fields acting on the sample create within its volume homogeneous magnetizations. This demands the fulfillment of two conditions: (1) the sample must be of such a shape that its demagnetizing field is homogeneous (sphere, ellipsoid, thin plate), and (2) it must be placed in those regions of the resonant cavity where there are no gradients of the high frequency magnetic field. The physical meaning of these conditions is clear: when they are fulfilled all the spins in the sample precess with the same phase. The violation of either of these conditions must lead to the appearance of inhomogeneities either in the constant or in the high frequency magnetizations of the substance under investigation and to a considerable complication of the picture of resonance absorption. These complications manifest themselves in the asymmetry of the observed resonance curve, in its broadening, and in the splitting of the resonance line into several lines of different width and intensity. The last effect which was observed comparatively recently received the not very appropriate name of multiplet ferromagnetic resonance.

The features indicated above do not fall within the framework of the usual theory,[1] and may be understood only on the basis of additional assumptions. In particular, the physical nature of multiplet ferromagnetic resonance is associated with the concept of the existence within the magnetized ferromagnetic medium of a spectrum of spin waves. As is well known,[2] the essence of this concept consists of the following. If the ferromagnet is placed into an external magnetic field, the small but unavoidable inhomogeneities of magnetization can be represented in the form of a superposition of spin waves propagated at a certain angle with respect to the direction of the field. In an infinite medium the spin waves are plane and form a continuous spectrum, while in the case of a sample of finite size the waves are described by more complicated functions (for example, in the case of a spheroid by Legendre functions) and form a discrete spectrum — the totality of the different vibration modes. Each mode corresponds to a certain eigenfrequency and to a certain structure of the magnetic field, and, moreover, cases of degeneracy are possible when the same frequency corresponds to several different types of oscillations which differ in their field structure.

Among all possible types of oscillation of a given sample there exists at least one whose frequency coincides with the frequency of the "normal" resonance given by Kittel's formula.* For large wave numbers (high frequency oscillations) the solutions obtained for an infinite medium may be approximately extended to the case of a sample of finite size. The practically important case of long spin waves is arrived at by the simultaneous solution of the equations of motion and of Maxwell equations without taking into account the exchange interaction and the propagation phenomena. This "magnetostatic" case was discussed by Walker[3] who calculated the low frequency spectrum of the characteristic oscillations of a spheroid.

The action of an external high-frequency field of appropriate structure on a ferromagnetic sample with inhomogeneous magnetization should result in resonance absorption at several frequencies, or, if the frequency is constant, at different values

*Cases are possible when Kittel's resonance does not occur.

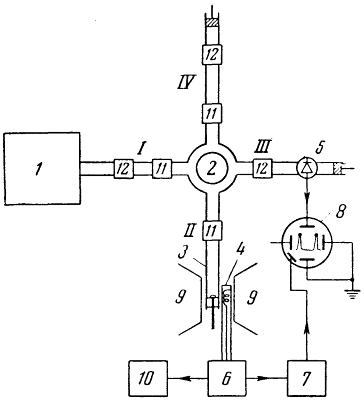

FIG. 1. Diagram of the apparatus: 1 – microwave generator; 2 – separating ring; 3 – segment of wave guide with piston and sample; 4 – fluxmeter coil; 5 – detector; 6 – fluxmeter; 7 – audio-frequency amplifier; 8 – oscillograph; 9 – electromagnet; 10 – fluxmeter wave meter; 11 – matching devices; 12 – ferrite isolators.

of the constant magnetic field. Such phenomena have been observed in specially designed experiments,[4,5] and have been partially studied.

Conditions necessary for such observations are clear from the foregoing: the high-frequency magnetic field of appropriate structure may be produced in the sample by violating either one of the conditions noted above necessary for the observation of "normal" resonance. Moreover, depending on the structure of the oscillating field set up in the sample the resonance absorption spectrum, in general, will be different all other conditions remaining the same.

In the papers quoted above the field of required symmetry was produced by violating the second of the foregoing conditions; the ferrite sphere[4] or disc[5] were placed in regions of the resonance cavity with the largest gradients of the microwave magnetic field. In the experiments described here the greater part of the observations was carried out with the first condition violated: the inhomogeneity of the high-frequency magnetization was produced by the inhomogeneity of the demagnetizing field of the samples.

Undoubtedly the interpretation of the results in the second case is much more difficult than in the first. However, having in mind the importance of the phenomena of multiplet ferromagnetic resonance both from the point of view of the physics of these phenomena in ferrites, and also from the point of view of many technical applications (first of all for the construction of a ferrite microwave amplifier) it appears to us to be useful to study all possible features of this phenomenon under various conditions.

2. EXPERIMENTAL PROCEDURE

The experiments were performed principally at a frequency $\nu_1 = 9200$ Mcs, and partially at $\nu_2 = 14640$ Mcs, by means of usual videospectroscopes[6,7] with a transmitting (at the ν_1 frequency) and a reflecting (at the ν_2 frequency) resonator at room temperature; some observations were taken at liquid-nitrogen temperature.

For sufficiently large samples, whose dimensions unavoidably could not be made smaller, we have tried a method of measurement which does not require continuous tuning associated with the frequency drift of the resonator containing the spheroidal sample. This circuit (Fig. 1) is based on the use of a separating ring which is the analogue of a double-T junction. Power from a stabilized klystron generator is brought through arm I to the separating ring in which it is split in half and enters arms II and IV, and if all the arms leading to the ring are matched it does not reach arm III. Arm II is connected to a section of a waveguide terminated by a shorting piston upon which the sample is mounted. The power reflected from arm II is divided between arms I and III; to the latter is connected a tune detector. Ferrite isolators mounted in arms I, III, IV decouple the generator from the ring and do not allow power reflected from arms III and IV to reach the generator. The system described above is equivalent to a semi-infinite line having the property that half of the power reflected from the load is brought to the detector and does not go back into the line. The system has lower sensitivity than a videospectroscope with a resonator but, as is shown by comparative measurements, the sensitivity is reduced by an amount which is insufficient to prevent the recording of even weak resonance signals from a strongly absorbing sample.

The constant magnetic field of intensity up to

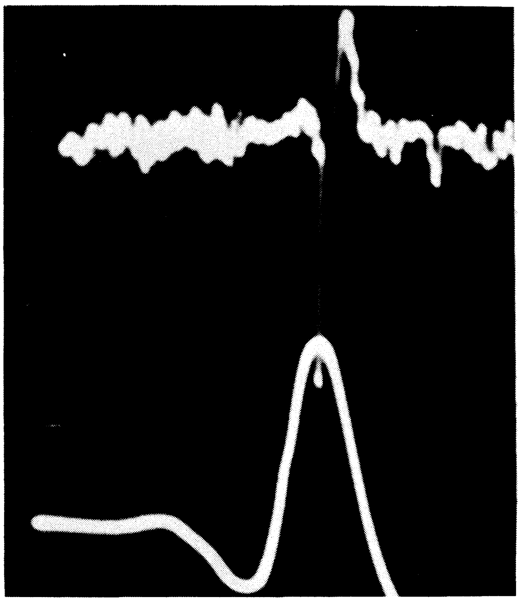

FIG. 2. Measurement of line position and width by means of a double beam oscillograph; lower beam-ferromagnetic resonance signal, upper beam-proton resonance signal in the same magnetic field.

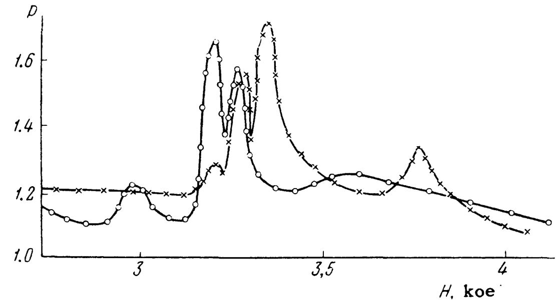

FIG. 3. Absorption by a MnZn polycrystalline ferrite sphere in positions: o – II and x – IV.

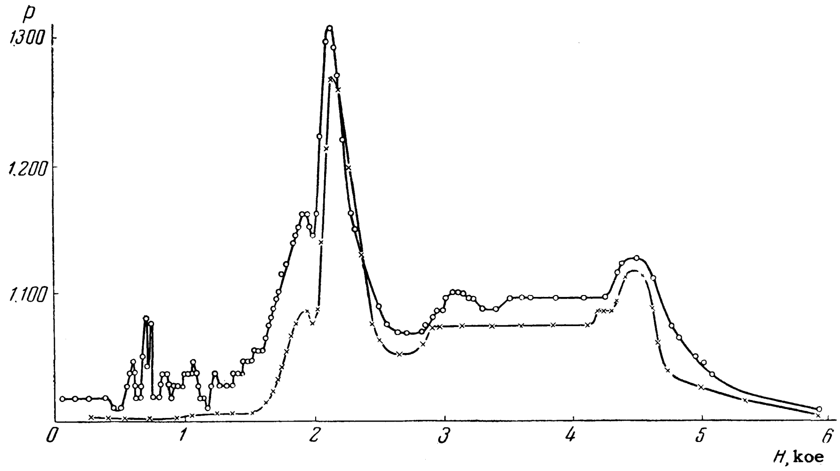

FIG. 4. Absorption by a polycrystalline ferrite ring in positions: x – I and o – III.

9000 oe was produced by an electromagnet with a pole diameter of 160 mm and a variable gap from 20 to 50 mm. The magnetic field was modulated with an amplitude up to 300 oe at 50 cps. The position of resonance absorption signals was determined by means of a proton fluxmeter whose coil was placed in the field of the same intensity as the sample. The proton resonance signal was applied either to the intensity control of a single beam oscillograph, or to the second beam of a double beam oscillograph. In the latter case the method of measurement of the position of resonance signals in the field and of their width is clear from the appended photograph (Fig. 2). The absorption curves were recorded point by point with continuous monitoring of the nature of the signals by means of the oscillograph.

Observations were made on the following samples: (a) single crystals of MnMg-ferrite in the shape of a hemisphere of diameter from 2 to 5 mm; (b) polycrystalline Mn, MnZn, and MnMg-ferrites with different impurities in the shape of rings of external diameter D from 2 to 5 mm, internal diameter d from 1 to 3 mm and of height h from 1 to 3 mm. The different ferrite samples had closely similar properties: a rectangular hysteresis loop, $H_c = 0.1 - 1.5$ oe, B_r from 1900 to 2500 gauss and B_m from 2000 to 2700 gauss.

In order to compare the results with those of other authors, we made several measurements using 2-millimeter spheres of polycrystalline MnZn-ferrite.

The samples were placed in three positions: position I — on the resonator axis at a distance of $\lambda/2$ from the shorting piston, this position corresponds to the condition of observation of "normal" resonance; position II — near the narrow wall at a distance of $\lambda/2$ from the piston; position III — on the shorting piston at its center. Moreover, in the case of spherical ferrites the spectrum was observed in position IV on the resonator axis at a distance of $\lambda/4$ from the piston. In positions I, III and IV the sample could be rotated about the resonator axis through any arbitrary angle which could be measured with an accuracy of the order of 10′.

3. RESULTS OF OBSERVATION

Figure 3 shows sections of microwave absorption curves at frequency ν_1 for a MnZn-ferrite sphere of 1.90 mm diameter in positions II and IV respectively in the range of fields from 2.8 to 4.0 koe. Both curves are very similar to the curves of White and Solt[4] taken at the same positions. The difference consists only in a certain displacement of the maxima towards weaker fields which, undoubtedly, is associated with the difference in the value of B_m, and in the less smooth shape of our curves. In addition to the principal maxima appearing in both curves at fields from 1100 to 7500 oe there are groups of considerably weaker maxima of different intensity and width also in agreement with the results quoted earlier. There does not seem to be any particular ordering in the position of the weak signals. Following Walker,[3] who has examined the results of White and Solt, the maxima in the curves of Fig. 4 may be explained as resonances of the long wavelength modes: (1,2,0) and (2,2,0) in the first case and (1,3,0) and (0,2,0) in the second case.

Both in the case of polycrystalline and of single crystal samples which differ in shape from spheroids the most complicated spectra are obtained in position III. Figure 4 shows absorption curves for a 3-millimeter ring of MnMg-ferrite ($H_c = 0.25$; $B_r = 1900$; $B_m = 2000$) at frequency ν_1 taken in positions I and III. We can see the similarity of the two curves in fields > 1900 oe: the three most

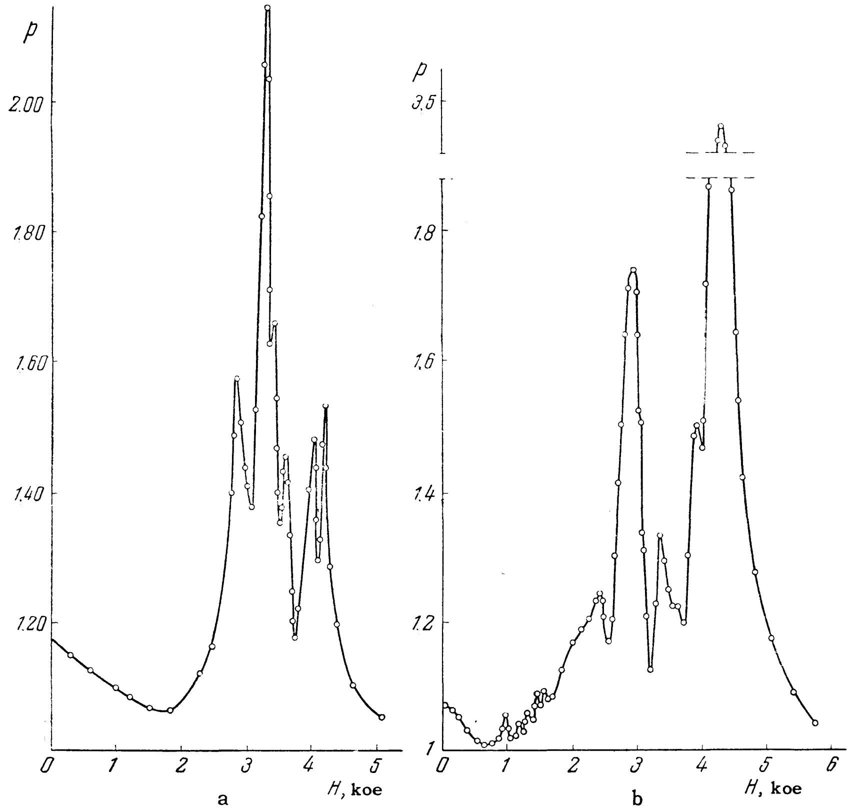

FIG. 5. Absorption by a MnMg-single crystal: a – in position II, b – in position III.

prominent maxima are preserved in going over from position I to position III practically without any change in position, and only with a slight change in intensity and width. However, in fields < 1900 oe the curves are quite different: in position III we observe a large number of less intense and much narrower absorption maxima, which do not appear in position I. Another characteristic feature is the appearance on this curve of a number of short horizontal stretches, which sometimes form a sort of a ladder, along which even with the most careful observations using a sensitive radiospectroscope it is not possible to observe any extremal points.

Figure 5 shows absorption curves for a MnMg-ferrite single crystal (hemisphere of diameter ~4 mm) taken in positions II and III. Here we find large differences both in weak and in strong fields. In fields < 2000 oe these differences are similar to those which occur in the case of a polycrystalline sample: in position III narrower and less intense signals appear which are not observed in position II.

The difference in absorption curves for the same sample placed at different points of the resonator volume are due to the difference in structure of the microwave magnetic field excited in the sample. In position I the characteristic structure of this field is due to the inhomogeneous demagnetizing field of the sample; in position II there is added to this the inhomogeneity of the magnetic component of the microwave field in the given region of the resonator; in position III the screening action of the conducting surface of the piston becomes evident which does not permit the flux of the microwave magnetic field outside the sample to complete its path in the same symmetric manner as in position I, which leads to a redistribution of the microwave magnetization inside the sample and produces a more complicated inhomogeneity in the magnetization, and thereby a structure of the microwave field which is essentially different from the structure in position I. It is natural to assume that in such cases the solution of the magnetostatic problem is considerably more complicated than the one in the cases considered in reference 3, if it is at all possible.

In the curves of Fig. 4 and 5b we can see a small number of additional signals appearing in position III in weak fields; actually, if observations are made with an oscillograph, a considerably greater number of weak signals is observed (in some cases up to 50 or so) and they occur at fields up to the order of 8 koe, however, the most prominent ones among them occur in the range from 400 to 1500 oe. In positions other than III a large number of weak signals is also observed but they occur in fields of not less than 1200 — 1500 oe

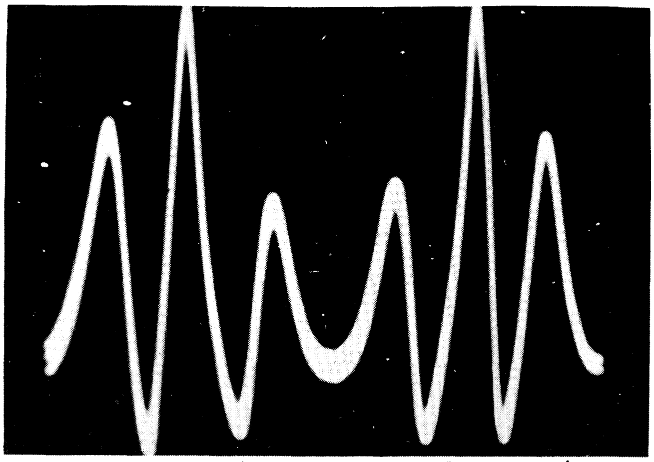

FIG. 6. Ferromagnetic resonance signals in a single crystal in a field of 585 oe (position III).

and, as a rule, are less intense than those observed in position III. An example of an oscillogram recorded in this position is given in Fig. 6. It gives us an idea of the shape, width and relative intensity of some of these signals. The widths of the signals fluctuate from 15 to 100 oe, while the intensity of the signals exceeds noise by a factor of from ~ 1 to 40. We did not succeed in noting any ordering in the position, width or intensity of these signals, other than the fact that separate groups are formed containing 3 — 8 signals (rarely more) which differ somewhat in their characteristics.

The position and the other characteristics of weak signals are quite different for different samples which are the same with respect to their composition, working, shape, and size. It can be asserted that the details of the "microstructure" of an absorption spectrum (particularly in weak fields < 2000 oe) are of an individual nature, and are not reproduced from sample to sample, although for the same sample placed under the same conditions the spectrum is very reliably reproducible. It appears that this provides a basis for the assertion that a considerable part of the weak signals of an absorption spectrum is due to the small inhomogeneities in the magnetic structure of the ferrites, which apparently unavoidably exist in each sample, and which have an individual character even in the case of exactly the same technique of preparation and working of the sample.

The picture of resonance absorption both in single crystals and in polycrystalline samples is very sensitive to the orientation with respect to the acting fields. All the absorption lines of the MnMg single crystal (with the exception of a small number of very weak ones) exhibit practically the same angular dependence (Fig. 7) which is in agreement

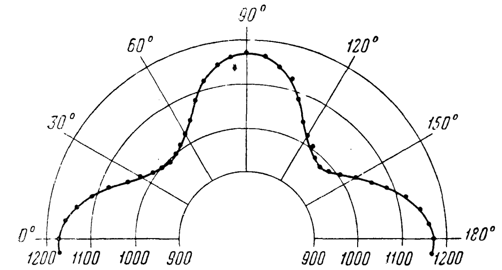

FIG. 7. Angular dependence of weak resonance signals in a single crystal. Rotation in the (100) plane; H in oe is plotted horizontally.

with theory and with previously known data obtained for the "normal" ferromagnetic resonance line.

The absorption lines of polycrystalline samples exhibit different angular dependence: while the fundamental signals like those shown in Fig. 5a do not depend on orientation, weak signals, particularly in fields < 1500 oe, are very sensitive to orientation. As the sample is rotated the signals move in the field, change their shape, width, and intensity, merge and again separate. Figure 8 presents the angular dependence of the position of two absorption signals in a ferrite ring, whose spectrum was given above, which are of interest due to the fact that at certain values of the field and angle they merge into one. The signals have a somewhat different angular dependence which is, however, in general characteristic of a single crystal. This suggests that such signals in a polycrystalline ferrite are due to local inhomogeneities situated at different depths below the surface, which have a different shape and which allow the setting up of vibrational modes of different structure. In the presence of strongly inhomogeneous demagnetizing fields in the sample the resonances of these modes may exhibit an angular dependence.

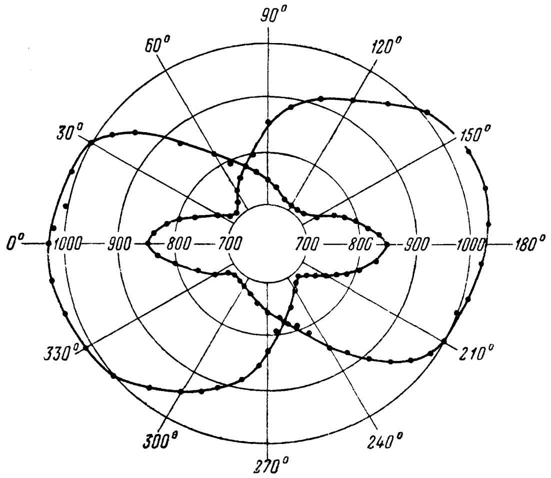

FIG. 8. Angular dependence of two weak resonance signals for a MnMg polycrystalline ferrite ring; H in oe is plotted horizontally.

It appears that the hypothesis that the weak resonance signals are due to local inhomogeneities within the volume of the sample is supported by the following experiment. A ferrite ring was set up in position III and was screened from above by a round very thin conducting plate of diameter equal to the diameter of the ring. We might have expected that the microwave field could penetrate only to a small depth below the external cylindrical surface of the ring, and that only those signals would survive on the absorption curve whose production was due to the inhomogeneity in the thin external layer of the sample. Indeed, only those signals remain on the absorption curve which appear in fields < 1600 oe and which do not change in shape or width. At the same time, signals in high fields (with the exception of a small number of very weak ones) disappear completely.

A few trial experiments at frequency ν_2 at room temperature and at liquid-nitrogen temperature using polycrystalline MnMg (with an admixture of ZnO) -ferrite ring (D = 1.06; d = 1.02; h = 1.00) in position III show that the intensity of weak signals varies but little with frequency. If the temperature is lowered to liquid nitrogen temperature the intensity increases by a factor of approximately 3 — 5.

The results enumerated above show that the phenomenon of multiplet ferromagnetic resonance is quite a complicated one depending on many conditions. Its further study may turn out to be useful both from the point of view of understanding the properties of ferrites, and also for technical applications.

The author expresses his sincere gratitude to A. I. Pil'shchikov for a number of very valuable discussions.

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