MAGNETIC MATERIALS

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MICROWAVE FERRITES

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Introduction

It is apparent from the title that this paper will be concerned with a limited region in the entire spectrum of ferrite applications. Unfortunately, the region is one which may present considerably less economic interest to the manufacturer of ferrite devices when the production quantities of ferrite microwave isolators are compared to those of ''fly-back'' transformers, for example. Perhaps, for this reason, together with the inherent problems of higher frequency technology, the rate of progress seems slow in achieving ultimate utilization of microwave ferrites.

This discussion will be concerned with a definition of primary loss mechanisms in ferrites, materials properties which are significant to the performance of typical microwave devices, current trends in ferrite applications, recent progress and problems in materials, and some observations regarding future ferrite development.

To simplify discussion, the microwave ferrites are grouped functionally in table 1, which relates also the significant effect with the type of device application. Any discussion of ferrite applications, especially those at microwave frequencies, requires some mention of dielectric and magnetic loss mechanisms.

Dielectric Losses

If we differentiate between true dielectric loss and loss due to high electrical conductivity, it is the latter which is most significant to ferrite performance. Conduction by mixed valences is, today, cited as the principal cause of conductive losses. This mechanism is typified by the existence of divalent and trivalent iron (Fe) ions in a ferrite which is deficient in oxygen because of composition or sintering processes. Ascribing losses primarily to mixed valence mechanisms may be a gross oversimplification in most cases. The divalent Fe in a ferrite being sintered may form a solid solution or displace, from the spinel lattice, other divalent ions which form a second phase. This happens with magnesium (Mg) ferrite (1), in which the divalent ion displaces Mg ions from the spinel lattice which form separate crystals of Mg oxide (periclase), and result in a very high conductivity ferrite. A look at the phase diagram of most ferrites indicates a narrow region of oxygen content over which the composition can contain only a single spinel phase as indicated in figure 1 for nickel ferrite ($NiFe_2O_4$). For the Mg-manganese system, there is no region at all in the vicinity of the stoichiometric composition where a single spinel phase may exist (2). Closer examination may reveal that many of the ferrites reported to have high or peculiar loss properties and behavior were beset by polyphase problems which made either analysis or correction of the deficiencies extremely difficult. Some of the means employed to prevent the effects of oxygen loss have been:

1. Sintering in oxygen at the decomposition pressure of the ferrite or in other gases.

2. Use of additives such as copper (Cu) and vanadium (V) to reduce sintering time and/or temperatures.

3. Introduction of a small amount of an ambivalent ion such as Mg or cobalt (Co) which is more readily reduced at high temperature than is trivalent Fe.

4. Replacement of appreciable amounts of the Fe with another element such as aluminum (Al), which exists only in the trivalent state. This affects the resultant ferrite in several ways; it increases the lattice stability, decreases the available oxygen which can be lost during sintering, and introduces interfering ions in the easy conducting paths which may otherwise exist.

Magnetic Losses

In ferrites, there is the familiar domain structure in which the elementary dipoles are all aligned and adjacent domains are separated by boundaries (termed Bloch walls) in which the spins gradually change direction. The wall has a thickness dependent upon conditions for minimizing the damping associated with spin exchange energy and the anisotropy. Magnetization of a ferrite results in alignment of the magnetic vectors with the applied field and growth of those domains favorably oriented with respect to the field. The effective permeability of a ferrite is the result of both effects, domain wall movement, and domain rotation.

Pringle (1) gives the following treatment for ferrites at microwave frequencies:

The initial permeability, μ_i, has real and imaginary components due to damping forces and is usually expressed as

$$\mu_i = \mu_i' - j\mu_i''$$

Where damping is small at low frequencies ($\mu_i'' = 0$) we can write:

$$\mu_i - 1 = 4\pi M_s^2 \left(\frac{1}{ad} + \frac{2}{3 M_s H_e} \right)$$

Here a and d represent constants involving the stiffness in domain wall motion and domain size, respectively, M_s is the saturation magnetization, and H_e is the effective internal field within a domain. Polder and Smit (3) show H_e to lie between the value of the anisotropy field, H_{anis}, and ($H_{anis} + 4\pi M_s$). H_{anis} is given by $4K_{1/3M_s}$ where K_1 is the first order crystalline anisotropy constant. Referring to the above equation, then, it can be seen that the first term represents the contribution due to domain wall movement and the second that due to domain rotation. Both processes lead to resonances in the μ_i'' frequency spectrum. Rado, et al. (4), gives the resonance angular frequency for domain wall movement as:

$$\omega_1 = \gamma (8\pi a)^{1/2} (A/K)^{1/4}$$

where γ is the gyromagnetic ratio and A the exchange interaction energy per unit volume. Snoek (5) shows that the resonance angular frequency for pure domain rotation (from $\omega_2 = H$) is

$$\omega_2 = \frac{8\pi\gamma M_s}{3(\mu_i - 1)}$$

where μ_i is the initial permeability at low frequencies.

"The condition for elimination of low field loss in an unsaturated ferrite medium is $f > f_{lim}$ where

$$f_{lim} = \frac{\gamma}{2\pi}\left(H_{anis} + 4\pi M_s\right)$$

For high field devices the limiting frequency is that at which the field required for resonance equals the saturation field, and this is intimately connected with ferrite geometry."

In general, the magnetic losses in devices employing modulating fields in the hundreds-of-kilocycle to tens-of-megacycle range can be categorized as primarily domain resonance losses at low fields and hysteresis losses at high fields (near saturation). Both of these arise from "stiffness" of the domains and domain walls. Hysteresis losses may be reduced by reducing anisotropy although the low field losses may remain high for a particular frequency due to resonance in domain rotation or wall motion. The anisotropy constant K_1 can be minimized by employing a ferrite with a low Curie Temperature such as the nickel-zinc ferrite (6), or by adjusting composition in a solid solution of two ferrites with anisotropy constants of opposite sign. Typical of this technique is the NiCo ferrite system (7) which uses a small amount of Co ferrite with a large positive K_1 in a Ni ferrite solid solution of a small negative K_1.

Low Field Devices

Devices based on the first effect, Faraday rotation (table 1), can be considered, in general, as low field devices. The low frequency limit in low field devices can be extended only by reducing both the saturation magnetization and the anisotropy (see the Polder and Smit relation, 3). One method of reducing the saturation magnetization is to substitute a nonmagnetic ion for part of the Fe (as in the ferrite aluminates).

High Field Devices

To eliminate low field loss in high field (resonance) devices, H_{sat} must be less than H_{res} and the condition for zero low field loss becomes $\omega > \gamma H_{anis}$. In a typical Ni ferrite with $\gamma = 3.22$ Mc/s/oersted and H_{anis} of about 400 oersted, the minimum frequency is about 1300 Mc/s. Lax (8) writes a relation for maximum reverse-to-forward ratio in a resonance isolator at a frequency, f, as

$$R = \frac{(4f)^2}{\gamma \Delta H}$$

which shows the importance of narrow line width at low frequencies for a given front-to-back ratio. Broadband applications require broad line width ferrites with fairly low γ, and narrow band applications with high back-to-front ratios require ferrites with narrow line width and low γ. Pringle reports X-band isolators with power ratios in excess of 120 to 1 using a ferrite such as $Ni_{0.975}Co_{0.025}Fe_2O_4$. More recently, ferrites such as Trans-

Tech414 have been used in X-band isolators and exhibit 20 to 40 db isolation (depending on power levels) with ½ to 1 db insertion loss, respectively.

In high power devices, we must account for nonlinear effects, as illustrated in figure 2, for a typical resonance isolator investigated by Pringle, which employs MgMn ferrite strips against the broad faces of a rectangular wave guide. Lowering and broadening of the resonance absorption peak and the appearance of a subsidiary absorption peak are explained by Suhl (9) as being excitation of higher order spin wave modes which grow exponentially at the expense of uniform precession when the rf field exceeds a certain threshold value. The threshold for saturation of the main resonance peak is given by

$$h_{crit} = \Delta H \sqrt{\frac{2\Delta H}{4\pi M_s}}$$

indicating the desirability of broad line width ferrites if these absorption anomalies are to be avoided.

High peak power applications thus require broad line width ferrites with a high Curie temperature (for high average powers); and, high power continuous wave (CW) applications indicate the additional need for high resistivities.

Intermediate Field Devices

Nonreciprocal phase shift is based on the interaction of the microwave magnetic field with the Larmor precession of the magnetization of a ferrite which is biased with a dc magnetic field. Figure 3 shows typical phase and attenuation characteristics plotted as a function of the dc field strength. Such nonreciprocal devices as isolators and circulators make use of the difference in propagation constant for positive and negative circular polarization. The variation in propagation with applied field is used in switches and modulators.

In nonreciprocal phase shift devices, the fields must be large enough to saturate the medium but must be well removed from the resonance peak. Ignoring low field losses, Lax gives a relation for the maximum figure of merit (in terms of differential phase shift-to-loss ratio) at a frequency, f, as

$$F_{max} = \frac{2f}{\gamma \Delta H}$$

At lower frequencies (figure 3), there is more overlap of the low field losses and resonance losses. The amount of overlap can be reduced by using material with a narrow resonance line width. Because of the low field losses, the figures of merit involving only line width are inaccurate below S-band, and ferrites for use in this range should also exhibit low saturation magnetization. In transverse devices operated above S-band, the ferrite is usually saturated so that resonance loss predominates; this makes a narrow line width of primary importance.

Devices Using Nonlinear Effects

In ferrimagnetic amplifiers and other devices based on nonlinear effects, narrow line width and high saturation magnetization are desirable to reduce pumping power. The narrow line widths are desirable in connection with mixers and doublers, depending on second-order terms in the time varying component of the magnetization vector. This implies the use of single crystal materials, and the inherent stability of the garnet structures would indicate greater ease in obtaining the needed higher resistivities.

Current Trends in Ferrite Applications

While the microwave properties of ferrites have been applied to a large number of useful devices, new and improved devices are vitally needed in several areas.

Considerable effort is currently being given to extending ferrite applications down to L-band and UHF. Availability of low noise microwave amplifiers has stimulated attempts to achieve lower insertion loss in conventional devices, particularly circulators. The advantages of inertialess antenna beam steering have aroused interest in the use of ferrites for rapid phase shifting or switching. Studies of high power effects indicate that ferrites may be useful for limiting, harmonic generation, and microwave delay techniques. Applications of ferrites and antiferromagnetic materials to millimeter wave devices are also being investigated.

Several requirements must be met by ferrite phase shifters before practical application of them can be made in electronically scanned antenna arrays. For a planar array, the phase shift taper across the array must be linear and must be an accurately known function of the applied signals from the scanning programer. Also, it must be possible to change the phase taper rapidly with a minimum of driving power. Successful application of ferrite phase shifters requires materials with very little variation of saturation magnetization with temperature. The requirement for uniform behavior of all the phase shifters in the array makes it desirable to use materials whose properties are closely reproducible from batch to batch. The difficulty of achieving this in polycrystalline ferrites would make it worthwhile to consider using long single crystals in this application.

For millimeter-wave devices operating at fields below resonance, a high saturation magnetization is the main requirement. For conventional ferrites, resonance at 70 KMC would occur for a field of approximately 25,000 oersteds, requiring an extemely bulky and power consuming room temperature magnet. Super-conducting magnets (10,11) would solve these problems, but the associated cryogenic equipment might be undesirable. The high anisotropy fields in certain ferrites can be used to aid the applied field, thereby alleviating the external magnet requirements. Barium ferrite has a uniaxial field of about 1700 oersteds, which adds directly so that resonance at 70 KMC requires an additional applied field of only 8000 oersteds (12,13). Antiferromagnetic materials also have high effective internal fields. Chromic oxide and manganese fluoride appear applicable to millimeter devices, although they must be used at liquid nitrogen and helium temperatures, respectively (14).

Harmonic generation of millimeter waves has been demonstrated using single crystal YIG spheres to obtain conversion efficiencies of a few percent for doubling from 70 to 140 KMC/s with input powers of a few kilowatts (15).

In addition to the nonlinear behavior mentioned in the preceding section, other high power effects occur which have application to millimeter-wave devices. The resonance broadening and appearance of a subsidiary absorption are illustrated in figure 2. For lower frequencies, coincidence of the main and subsidiary resonance occurs and and expression for critical power (16) may be written as:

$$P_{crit} = K \frac{(\Delta H)^4}{(M_s)^2}$$

At higher frequencies, the critical power is expressed by

$$P_{crit} = K \frac{(\Delta H)^3}{M_s}$$

This means that the critical power in single crystal YIG with M_s = 1780 gauss and ΔH of about one oersted may be a few microwatts at frequencies below 3300 MC/s and a few milliwatts at higher frequencies. Broadening of the main resonance results in reflection limiting in a magnetic resonance filter, and the appearance of the subsidiary resonance results in absorption limiting. While restricting the power handling capability of ferrite devices, this limiting may be used to advantage for stabilizing the output level of CW sources or for preventing saturation of CW amplifiers.

This high power effect has been described previously as due to excitation of higher order spin wave modes. The spin waves are coupled to phonons (17,18). Bommel and Dransfeld (19) used this fact to make a transducer between a microwave cavity and a quartz rod for propagation of acoustic waves at microwave frequencies. Availability of long single crystals of garnets might stimulate some experiments in the use of garnets as sonic delay elements at microwave frequencies.

Progress in Materials

The past year saw the first device which uses a single crystal ferrimagnetic oxide marketed. Using a concept suggested by DeGrasse, the Watkins-Johnson Company has developed a tunable filter operating over 2 to 4 KMC which uses a single crystal YIG sphere. Using the same concept, work on ferrimagnetic limiters is being done in many laboratories. The use of single crystal ferrites of higher saturation magnetization will permit higher frequency operation of both types of devices. Lithium ferrite is now the best candidate for the latter application.

The growth of single crystals of garnets has been further perfected. From a flux, Nielsen has obtained crystals (figure 4), which weigh up to 95 g each and are 80 percent sound. Kramarsky (not published) has grown crystals of YIG with ferromagnetic resonance line widths approaching 0.2 oersteds at room temperature.

Nothing representing a notable breakthrough has occurred recently in polycrystalline ferrite compositions. Progress has mostly been in the processing techniques where efforts have been made to minimize losses and ensure uniformity and reproducibility in ferrites.

The magnitude of the low field loss due to domain wall motion can be reduced by the use of fine-grained ferrites in which the crystallite size is less than the size of a single domain. However, it is still necessary to dilute the ferrite to prevent the crystallites from interaction which will cause a considerable increase in the line width.

One very promising improvement has been reported by Malinofsky (20). He has prepared Ni ferrite by the flame spray technique and has achieved particle sizes of less than the domain size of the material. The domain rotation loss observed in most ferrites near

1000 MC was completely suppressed. This result could be important to those microwave applications where low field losses are a problem.

Although not strictly in the materials area, the observation by Hartwick, Peressini, and Weiss (21) that the subsidiary resonance in YIG crystals can be completely suppressed by modulating the dc field is extremely important. This experiment was performed to test a prediction of Suhl's (22), which promises to increase the signal level at which ferrites can be used. Weiss (23) has also suggested that the proper modulation of the dc field may lead to early appearance of subsidiary resonance. This would make very low level limiters possible.

The problems in ferrites remain much the same, that is, poor reproducibility, poor loss characteristics, etc. In addition, the increasing use of high power levels in radar systems compound these difficulties. Possibly, there are still ferrite systems and processing techniques left which will drastically improve ceramic ferrite performance, but much work has been done and the performance of most compositions can be predicted fairly well. Thus, barring discovery of a new system, the power levels above which ferrites cannot be used are probably in sight. For instance, Schlomann, et al. (24), has suggested the upper limit of peak power for resonance isolators to be several megawatts.

The problem of making low loss, low saturation, high Curie temperature ferrites for S- and L-band devices has not been completely solved although gadolinium substituted YIG has come into use more and more.

In the single crystal growth field one big problem remains —the growth of sound, large crystals of ferrites and substituted garnets which are stoichiometric and of uniform composition. In other words, the problem of the growth of large, uniformly stoichiometric crystals of high melting oxide solid solutions has not yet been solved.

Conclusion

In the light of much of the past effort in developing ferrite materials and applications, there is a point which can hardly be overemphasized. Significant advances in any of these areas require not only efforts in developing ferrites with microwave properties tailored for particular applications, but also investigations of the physical mechanisms affecting the microwave properties. Recent years have brought significant advances in ferrite theory and a greater understanding of their behavior in microwave and other applications. Much of this resulted from the observations made in carefully conducted empirical investigations. However, a tremendous amount of effort devoted to experimentation or "development of improved ferrites" has been of dubious, if any, value. For example, some extensive programs were launched to establish quantitative correlation between processing parameters of some low loss ferrites before properly determining that a specific single phase material actually contained an extremely "lossy" second phase.

Recommendations

Some considerations for future effort in the area of ferrite materials are:

1. The effect of materials purity as well as oxygen stoichiometry on conductive behavior. This would involve a rather detailed treatment of conduction processes and a definition of constitutional diagrams to differentiate between single phase and "spinel-plus" compositions.

2. Preparation and study of single crystals in representative ferrite classes and serious attempts to bring polycrystalline properties nearer to those of the single crystal. Rupprecht (not published) has shown that losses in the ceramic form of $SrTi0_3$ dielectrics can be brought to within a factor of twice the single crystal properties by control of purity and post-sintering anneals.

3. Survey of ferrite properties over respectable temperature and frequency spectra. As indicated by some of the considerations in this discussion, a spot check of ferrite properties at a single frequency well below the intended frequency may be hopelessly inadequate in many cases. This may explain why published literature on microwave ferrites with property data of only 20 MC is so discouraging. Goodwin (not published) has conducted extensive measurements on a number of commerical ferrites and garnets at 77°K and at 4.2°K.

4. A first look at thiospinels or ferrites based on other unusual spinel systems.

5. Consideration of topotactical grain orientation, magnetic anneal, or other means for achieving orientation in ferrites for specific purposes.

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TABLE I

MICROWAVE FERRITE APPLICATIONS

FARADAY ROTATION	PHASE SHIFT	RESONANCE ABSORPTION	SECOND-ORDER EFFECTS	NON-UNIFORM SPIN WAVE MODES
Switch Circulator	Gyrator	Resonance Isolator	Frequency Doubler	Ferromagnetic Amplifier
45-Degree Rotational Isolator	Field Displacement Isolator	Circular Polarizer	Mixer Detector	Limiter
Amplitude Modulator	Frequency Modulator			

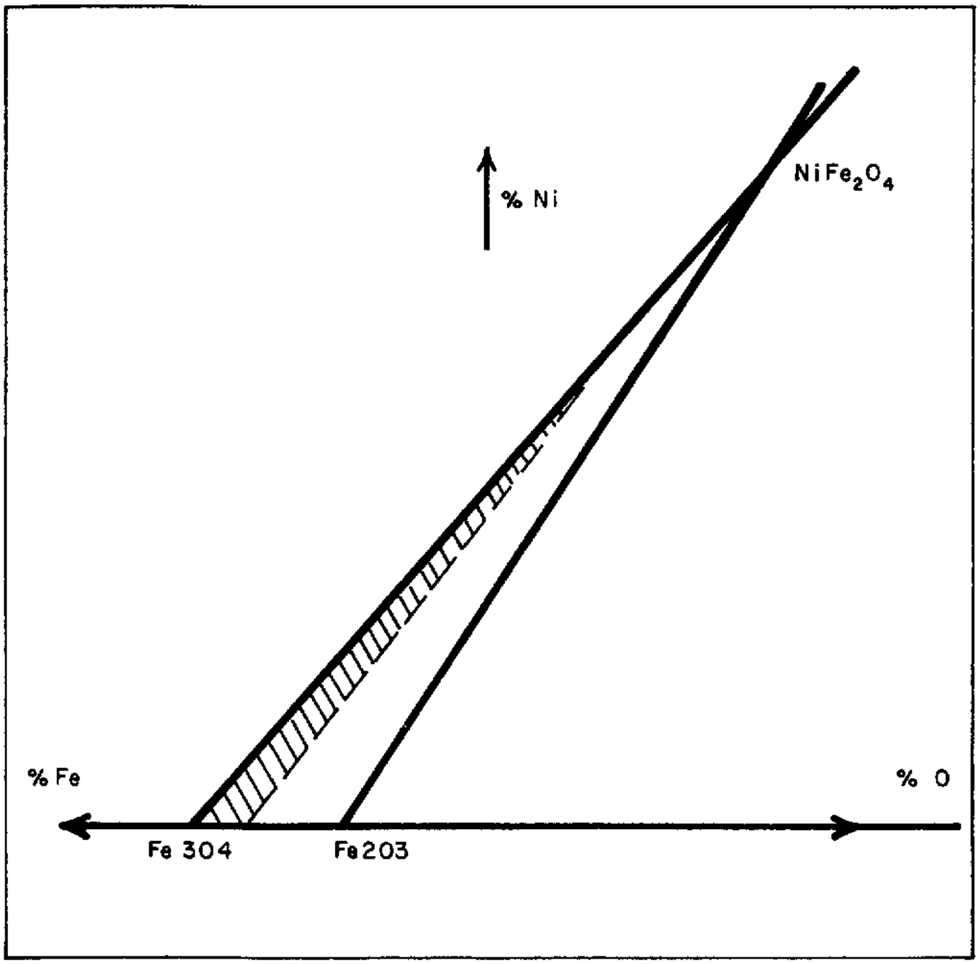

Figure 1. Monophase Spinel Region for Ni Ferrite

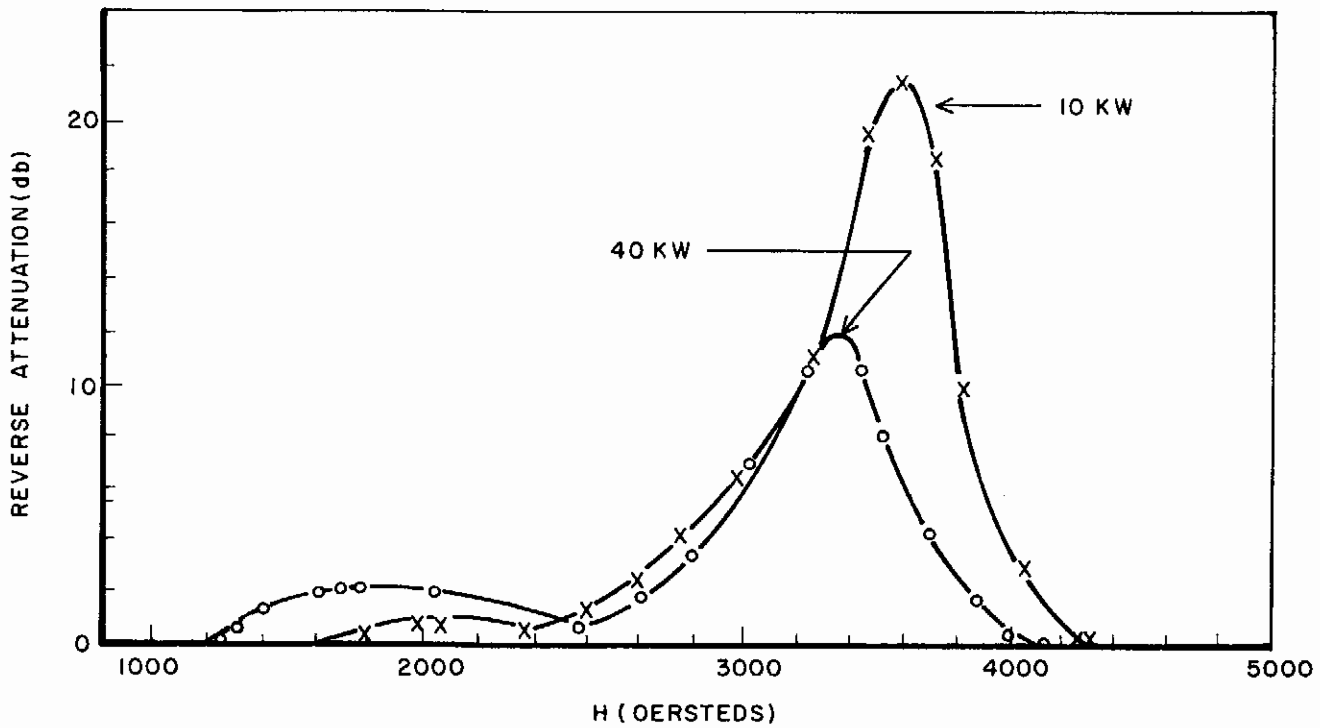

Figure 2. Effect of Power Level on Resonance Absorption of a Mg-Mn Ferrite

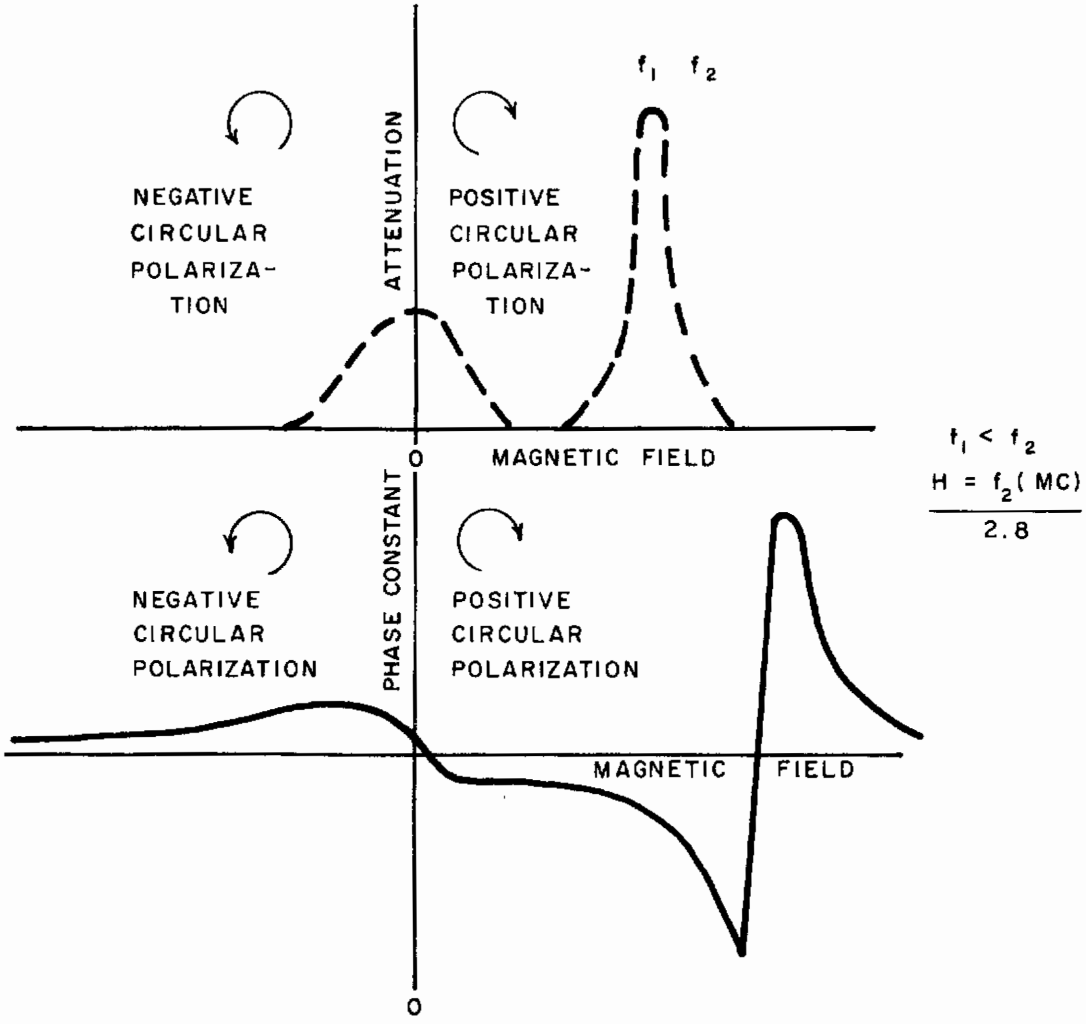

Figure 3. Typical Phase and Attenuation Characteristics Vs Applied Field

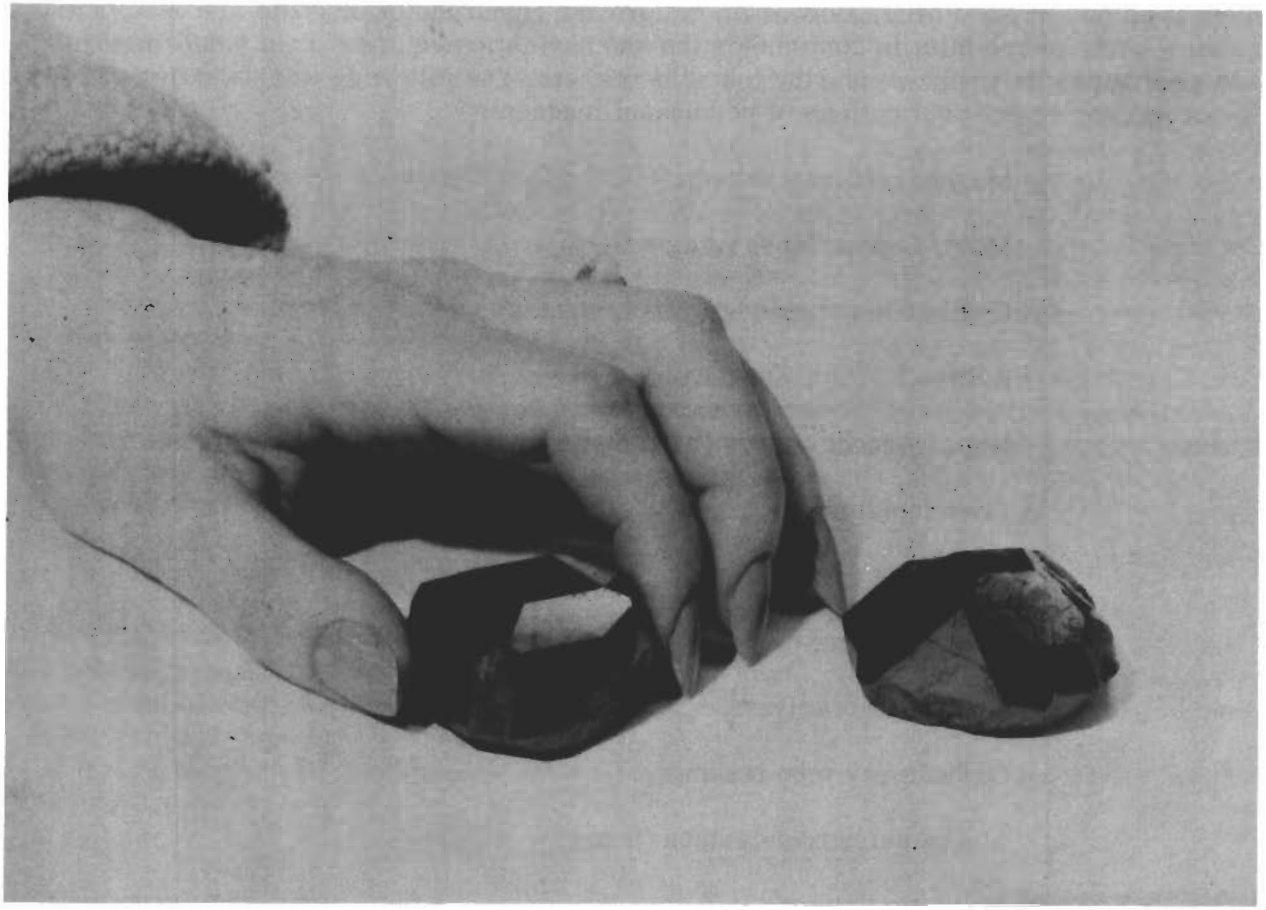

Figure 4. Single Crystals of Garnets