Microwave Lithium Ferrites: An Overview

GIL M. ARGENTINA AND PAUL D. BABA, MEMBER, IEEE

(Invited Paper)

***Abstract*—Lithium ferrites are discussed and compared with other spinel microwave ferrites and rare earth-iron garnets. Points of comparison are saturation magnetization, temperature performance, hysteresis loop properties, stress sensitivity, insertion loss, power handling capability, resonance linewidth, and cost. The main points of discussion deal with the relative effectiveness of lithium ferrites, nickel ferrites, magnesium ferrites, and garnets as elements employed in latching applications at frequencies in the S, C, X, and K_u bands.**

A section is devoted to the compositional modifications necessary for: 1) adjusting magnetization, spin-wave line width, coercive force, and magnetic anisotropy; 2) the minimization of stress sensitivity and dielectric loss; and 3) the improvement of microstructural characteristics.

I. INTRODUCTION

THE category of microwave ferrimagnetic materials can be divided into two principal types: the rare earth-iron garnets, and the cubic spinel ferrites. Though limited by a saturation magnetization ceiling of 1800 G, the garnets have been the most important type for many years. Yttrium-iron garnet (YIG) can be chemically modified to enhance practically every performance characteristic considered important by the device designer [1].

Spinels of the nickel and magnesium-ferrite systems have long been used by the microwave industry. Due to severe limitations in temperature performance or loss, spinel compositions of lower magnetizations have been virtually eliminated from use in S- and C-band latching applications.

Recent development efforts [2] have resulted in a lithium-ferrite system which offers performance advantages over the other spinels and is competitive with garnets for C-band latching applications.

II. DESIRABLE CHARACTERISTICS OF MICROWAVE FERRIMAGNETIC MATERIALS

A. Magnetization

For a ferrite system to be useful over the full range of microwave frequencies, the room temperature saturation magnetization must be adjustable from 200 to 5000 G to satisfy loss requirements, as will be discussed in what follows.

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The authors are with Ampex Corporation, Redwood City, Calif. 94063.

B. Loss

Insertion loss is composed of both dielectric and magnetic loss. Low field magnetic loss, which is the major consideration for latching applications, is the result of domain resonance [3], and/or relaxation phenomena [4]. Conditions for minimum magnetic loss can be defined by the relation

$$\frac{\omega_m}{\omega} = \frac{\gamma(H_A + 4\pi M_s)}{\omega} < 1 \tag{1}$$

where H_A and $4\pi M_s$ are the anisotropy field and saturation magnetization of the ferrimagnetic material; γ is the gyromagnetic ratio; and ω is the operating frequency. Many spinels have $H_A \sim 100$ Oe which results in a contribution to the magnetic loss according to (1). When $H_A \ll 4\pi M_s$, it may be ignored. In such cases, the condition for low loss becomes $\omega_m/\omega = \gamma 4\pi M_s/\omega < 1$. Materials which have high anisotropy require a corresponding reduction in $4\pi M_s$, which is costly in terms of phase shift. For good device performance, μ'', the imaginary part of the magnetic permeability tensor, should be $< 4 \times 10^{-3}$ [5].

Dielectric loss in ferrites is generally ascribed to conduction by means of electron hopping between the two iron ions, Fe^{+2} and Fe^{+3}. According to Schlömann, dielectric loss does not contribute to insertion loss as long as $\tan \delta_\epsilon < 3 \times 10^{-4}$ [6]; there is also considerable evidence that levels of $\tan \delta_\epsilon$ slightly greater than 5×10^{-4} can be tolerated with a minimal contribution to insertion loss.

C. Power Handling

The power handling capability of ferrimagnetic materials is limited by the onset of magnetic instabilities known as spin waves. Once excited, spin waves cause a nonlinear increase in loss.

Power handling varies directly with the spin-wave linewidth (ΔH_k) and inversely with the magnetization for a fixed frequency. The spin-wave linewidths of garnets and spinels can be broadened from 2 to 20 Oe to accommodate existing high-power design requirements.

D. Hysteresis Loop Properties

Remanent magnetization B_r is the most important of the B–H loop properties for latching applications. Phase shift varies in direct proportion with B_r, and since magnetization is limited by $\omega_m/\omega < 1$, the ratio of $B_r/4\pi M_s$ must be as high as possible. In many of the spinel ferrites and garnets, remanence ratios of 0.75 are obtainable.

For most phase shifter applications employing saturation drive techniques, temperature sensitivity of B_r should be $< \pm 2$ G/°C over the temperature range 0°C to +80°C. The gadolinium-doped yttrium-iron garnets (Gd YIG) used at S band have magnetic compensation points below room temperature, and as a result can have temperature sensitivities of $< \pm 1$ G/°C.

Due to the ferrite-to-waveguide contact experienced in most phase shifter designs, stress is encountered during normal temperature cycling. It has been demonstrated that the effect on B_r of uniaxial stress applied parallel to the direction of the magnetic field is predominantly determined by the ratio of the hard axis magnetostriction constant to the anisotropy constant [7]. The garnets are particularly stress sensitive. While Mn^{+3} additions [8] have appreciably reduced bending stress sensitivity in garnets, loads of ~500 lb/in² result in changes in B_r on the order of 10 percent. Magnesium and lithium ferrites are practically stress insensitive, with changes in B_r of less than 2 percent for the same test conditions.

Switching energy requirements for phase shifters is dependent on the coercive force and the switched flux of the ferrimagnetic element. Coercive forces of less than 1 Oe are small enough to permit the use of low-cost solid-state drivers.

E. Resonance Linewidth

Though of no particular importance in the design of latching devices, resonance linewidth is perhaps the most critical design consideration for above resonance devices such as circulators. The aluminum substituted YIG's are the most widely used materials for above resonance circulators. These materials have linewidths of ~45 Oe. Some of the lithium-ferrite spinels are close to the garnets in performance; however, the nickel and magnesium ferrites are not at all suitable for above resonance application due to broad resonance linewidths.

Recent efforts [9], [10] have resulted in calcium-vanadium garnets with extremely narrow linewidths (10–20 Oe).

III. BACKGROUND ON LITHIUM FERRITES

Lithium ferrites became commercially important as computer memory core materials in the early 1960's. The high Curie temperature, leading to unparalleled thermal stability, the excellent hysteresis loop properties, and the high saturation magnetization all prompted this commercial interest. For many of the same reasons there was considerable development effort aimed at providing microwave quality lithium ferrites [11]–[14].

The principal interest in microwave lithium ferrites is as a low-cost replacement for the rare earth-iron garnets, offering competitive or improved temperature performance. Lithium ferrites with magnetizations comparable to the garnets are very refractory due to the high concentration and nature of the substituent elements, which requires relatively high sintering temperatures. This type of heat treatment causes the volatility of Li_2O [15], [16] which results in some reduction of iron. For this reason lithium ferrites were considered difficult to prepare with low dielectric loss. In addition, these lower magnetization lithium ferrites were characterized by high porosities, and as a result, high coercive forces and broad resonance linewidths were experienced.

IV. COMPOSITIONAL CONSIDERATIONS FOR EFFECTIVE MICROWAVE LITHIUM FERRITES

A. Reduction and Elevation of $4\pi M_s$

Unsubstituted lithium ferrite has a saturation magnetization of ca. 3700 G. In order to tailor this material to specific microwave requirements, compositions must be prepared with room temperature magnetizations from 200 to 5000 G.

Lithium ferrite has the formula

$$(Fe^{+3})[Li_{0.5}^{+}Fe_{1.5}^{+3}]O_4. \qquad (2)$$

The parentheses ( ) and the brackets [ ] indicate ions distributed on tetrahedral and octahedral sites, respectively. The magnetic spins on each of the sites are antiparallel; so, the octahedral site is dominant. For reduction of magnetization a nonmagnetic ion (or a magnetic ion having fewer spins than Fe^{+3}) is introduced on the octahedral site as a replacement for Fe^{+3}. To increase the magnetization, a nonmagnetic ion is introduced on the tetrahedral site. In either case, the octahedral site normally remains dominant.

Ionic substitution must be performed in a manner which insures electronic charge neutrality. For example, Al^{+3} substitutes equivalently for Fe^{+3}. Divalent (+2) and quadrivalent (+4) ions require additional adjustment. These substitutions can be accomplished by the following formulas:

$$\text{divalent } (+2) \qquad Li_{0.5-0.5x}^{+}Fe_{2.5-0.5x}^{+3}M_x^{+2}O_4 \qquad (3)$$

$$\text{trivalent } (+3) \qquad Li_{0.5}^{+}Fe_{2.5-x}^{+3}M_x^{+3}O_4 \qquad (4)$$

$$\text{quadrivalent } (+4) \qquad Li_{0.5+0.5x}^{+}Fe_{2.5-1.5x}^{+3}M_x^{+4}O_4. \qquad (5)$$

The specific substituent ion is selected on the basis of ionic size and crystal field, which influence site preference and the chemical stability of the ion. Ti^{+4} and Al^{+3} are nonmagnetic ions which have very strong octahedral site preferences and work very well for magnetization reduction. Ti^{+4} has been used by several investigators [2], [11], [13], [14], [20]. Al^{+3} is very difficult to work with in that it forms an immiscible compound with Li and O, under normal processing conditions [16], over much of

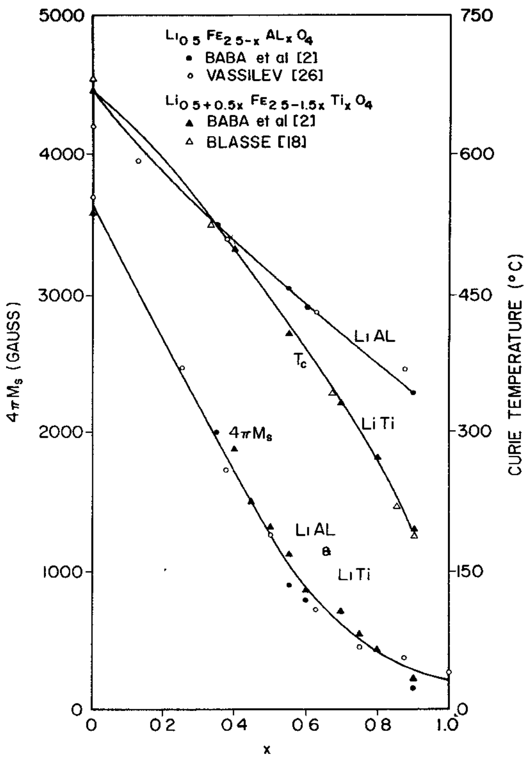

Fig. 1. Room temperature saturation magnetization and Curie temperature as a function of Al and Ti substitution in lithium ferrite.

the compositional range. The nonmagnetic ion Zn^{+2} has a very strong tetrahedral site preference and is commonly used for elevation of magnetization in all spinel-ferrite systems.

The only drawback to the use of a quadrivalent ion such as Ti^{+4} is that it requires the addition of some Li^{+} [see (5)] which is believed to occupy the tetrahedral sites [18]. This weakens the exchange interaction between the sites. As a result, a lithium-titanium ferrite does not have as high a Curie temperature as does a lithium-aluminum ferrite of equivalent magnetization. This effect can be seen in Fig. 1.

B. Sintering Aids

Lithium ferrite is difficult to sinter to high density. The substitution of Ti^{+4} to effect a lowering of $4\pi M_s$ increases this refractory nature. Temperatures in excess of 1200°C are required to sinter a typical 1000-G $4\pi M_s$ lithium-titanium ferrite to a density of around 90 percent of theoretical maximum. At these temperatures the loss of lithium, oxygen, or both [14], [15] is reflected by high dielectric loss, the result of the irreversible reduction of some Fe^{+3} to Fe^{+2}.

To alleviate this problem it is necessary to improve the chemical reactivity of the system. Several methods have been proposed, some of which employ highly reactive raw materials; for example, fine particle Fe_2O_3 and molten salts [19]; however, the most practical method to insure high density concomitant with a low sintering temperature is to employ a sintering aid. These materials are of two principal types: those which work in the solid state by promoting crystallographic defects that enhance diffusion, and those which form a liquid phase which increases diffusion rates. The latter method, known as liquid phase sintering, has been employed successfully by Baba *et al.* [2] using Bi_2O_3 as the fluxing agent in the sintering of lithium ferrites. Fig. 2 shows the effect of trace amounts of Bi on the sintering behavior of a lithium-titanium ferrite. It can be seen that densities of ~99 percent of the theoretical maximum can be achieved at 1000°C. The same composition without Bi has a density of ~75 percent of theoretical at the same sintering temperature.

The use of a sintering aid has the attendant advantage of reducing the coercive force. This is accomplished by the reduction of porosity and the increased grain size experienced through liquid phase sintering. Fig. 3 shows

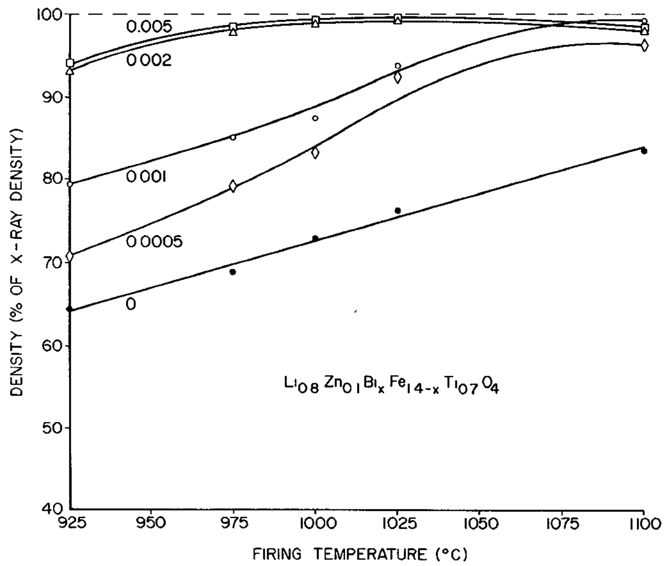

Fig. 2. Density versus sintering temperature and bismuth content for the composition $Li_{0.8}Zn_{0.1}Bi_xFe_{1.4-x}Ti_{0.7}O_4$.

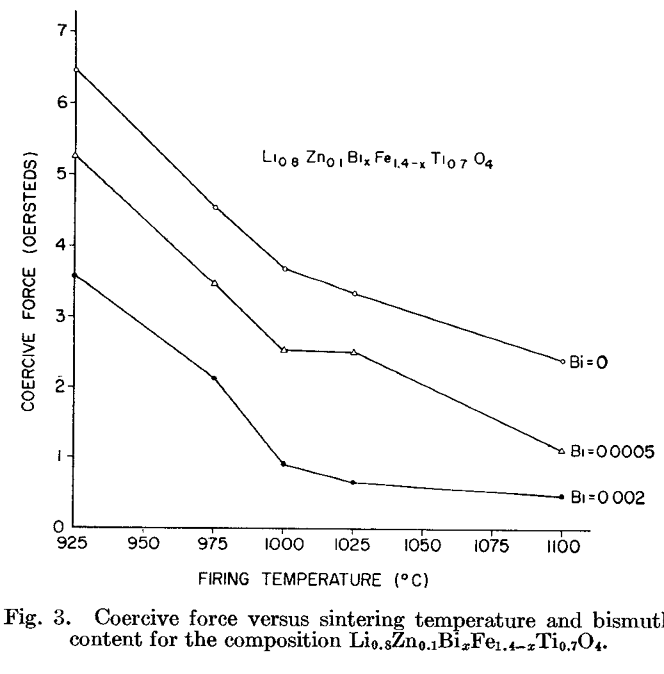

Fig. 3. Coercive force versus sintering temperature and bismuth content for the composition $Li_{0.8}Zn_{0.1}Bi_xFe_{1.4-x}Ti_{0.7}O_4$.

the effect of Bi on the coercive force of a lithium-titanium ferrite. A coercive force of <1 Oe can result at sintering temperatures of $\sim$1000°C.

C. Reduction of Magnetic Anisotropy

Lithium-titanium ferrites are relatively high anisotropy materials. The high anisotropy adversely effects low field magnetic loss in low magnetization (<800 G) ferrites and broadens resonance linewidth.

Effective reduction of anisotropy can be accomplished by the addition of Zn^{+2}. Fig. 4 shows this effect on series of 400-G $4\pi M_s$ lithium-titanium ferrites. This is not accomplished without sacrifice, however, and the Curie temperature for this series decreases from 220°C for 0 Zn^{+2} to 120°C for the composition with 0.15 ions per formula unit of Zn^{+2}.

D. Variation in Spin-Wave Linewidth

To be effective as a microwave ferrite system, lithium ferrites must offer a variability in power handling capability. This can be accomplished by variation in the spin-wave linewidth. Doping with small quantities of Co^{+2}, a fast relaxing magnetic ion, allows for continuous alteration of spin-wave linewidth from 2 Oe to $\sim$8 Oe, as is shown in Fig. 5.

The efficacy of Co^{+2} in increasing ΔH_k in lithium-titanium ferrites is well known [2], [20], [21]. Green [21] has reported a ΔH_k of 13.7 Oe in a Co^{+2} doped lithium-titanium ferrite.

E. Minimization of Dielectric Loss

Trivalent manganese doping has been employed to minimize dielectric loss in a variety of spinel ferrites. It has been reported that this is accomplished by a simple oxidation–reduction reaction with Fe^{+2} [22], [23]. Mn^{+3} works very well in the lithium-ferrite system for minimization of $\tan \delta_\epsilon$ to $< 5 \times 10^{-4}$ with the added advantage of reducing the magnetostriction constants [24].

V. MATERIALS SYSTEMS OVERVIEW

A comparison of commercially available garnets and ferrites for phase shifter applications is made.

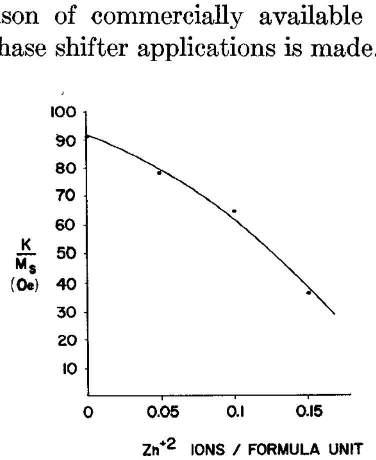

Fig. 4. Anisotropy versus zinc concentration in 400-G $4\pi M_s$ lithium-titanium ferrites.

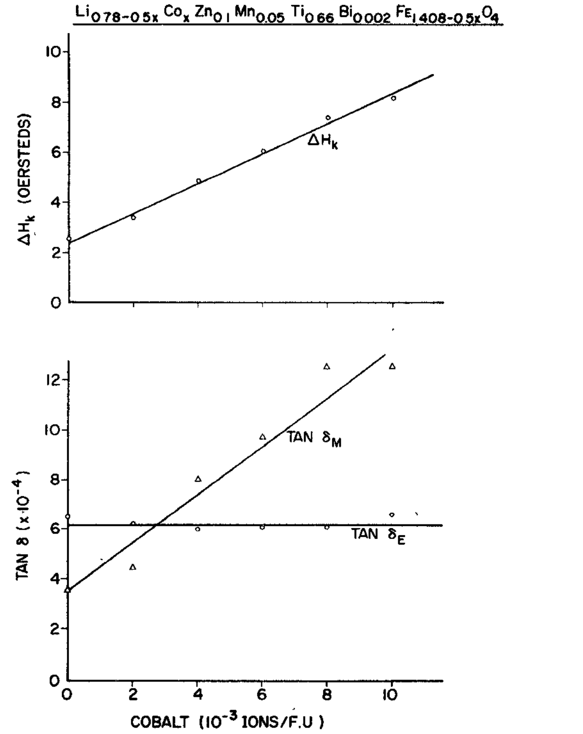

Fig. 5. ΔH_K, $\tan \delta_m$, and $\tan \delta_E$ versus cobalt concentration for the composition $Li_{0.78-.05x}Co_xZn_{0.1}Mn_{0.05}Ti_{0.66}Bi_{0.002}Fe_{1.408-0.5x}O_4$.

A. Magnetization and Temperature Performance

Fig. 6 depicts $4\pi M_s$ versus Curie temperature for the Mg, Ni, and Li spinels and the Y Gd Fe garnets (Gd YIG's). Two types of lithium ferrites are shown, varying in anisotropy. The temperature performance of the Gd YIG's which are magnetically compensated and

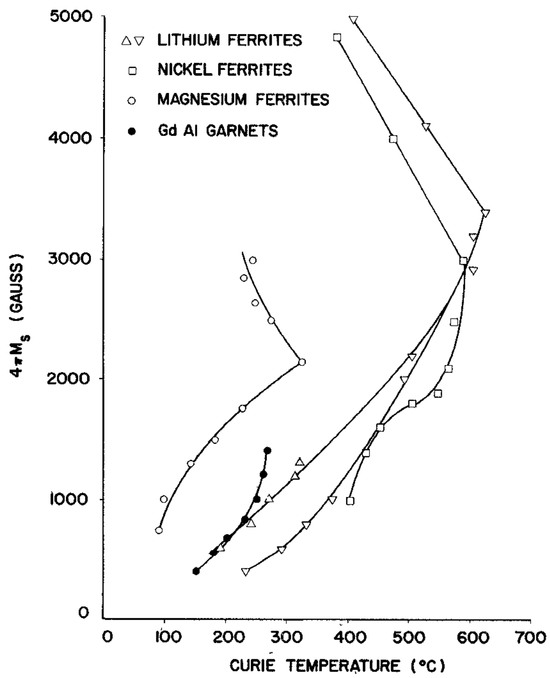

Fig. 6. $4\pi M_s$ versus Curie temperature for nickel, magnesium, and lithium ferrites, and gadolinium-aluminum YIG's.

have excellent temperature stability cannot be judged by this criterion; for these materials Fig. 6 serves only to demonstrate the limited range of magnetizations possible with the garnet system.

Figs. 7–10 show the temperature dependency of the remanent magnetization for optimum compositions for application at S, C, X, and K_u bands. The parameter B_r percent shown in the figures is defined as

$$B_r \text{ percent} = \frac{B_{r(-50^\circ \text{C})} - B_{r(125^\circ \text{C})}}{B_{r(-50^\circ \text{C})}}. \quad (6)$$

Two lithium ferrites are shown in Fig. 7, again varying in anisotropy.

B. Hysteresis Loop Properties

With the exception of the nickel ferrites, all of the materials under discussion with $4\pi M_s < 2000$ G can be processed to yield excellent hysteresis loop properties. The lithium ferrites and garnets, in nearly all cases, are available with remanence ratios $(B_r/4\pi M_s)$ of approximately 0.75 with the lithium ferrites generally enjoying a small advantage. Fig. 11 showing B_r versus $4\pi M_s$ for the spinels is of particular interest in the $4\pi M_s > 2000$ G region. Here improvements on the order of 15–50 percent in remanence ratio are obtainable with the lithium-ferrite system in comparison with other spinel ferrites.

Coercive force for magnesium and lithium spinels and garnets are low enough to avoid switching energy problems. The nickel ferrites have relatively high H_c.

C. Insertion Loss and Power Handling Capability

In the range of magnetizations 200 to 800 G, the lithium ferrites with the best temperature performance have inherently high anisotropy fields. This results in higher magnetic loss than a Gd Al YIG of the same magnetization (<800 G). Ordinary methods for reduction of anisotropy in the lithium-ferrite system increase the temperature sensitivity. In referring to Fig. 7, curve 2 represents the performance of a lithium ferrite which is equivalent in loss to Gd Al YIG's of the same $4\pi M_s$. This particular lithium ferrite is too temperature sensitive for most saturation drive phase shifter applications. A recent design [25] employing this same material with flux drive has resulted in excellent phase shifter performance. The lithium ferrite shown in curve 3 of Fig. 7 enjoys temperature performance close to the garnet but due to higher anisotropy is lossier.

Both the Gd Al YIG's and lithium ferrites are available for high-power applications. At the present time, however, the Gd Al YIG's offer superior temperature stability at S band for equivalent loss, magnetization, and power handling.

At C-band frequencies and above, the effect of anisotropy on low field magnetic loss is negligible. As a result the lithium ferrites and Gd YIG's are very close in every performance aspect.

Due to the requirement for $4\pi M_s > 2000$ G spinel ferrites are generally employed for X-band frequencies and above. At these higher frequencies, the lithium ferrites dominate with temperature stabilities equivalent to the Ni ferrites and with very low insertion losses comparable to the Mg ferrites. Lithium ferrites in the magnetization range of from 2000 to 5000 G are available with spin-wave linewidths, high enough to satisfy most requirements.

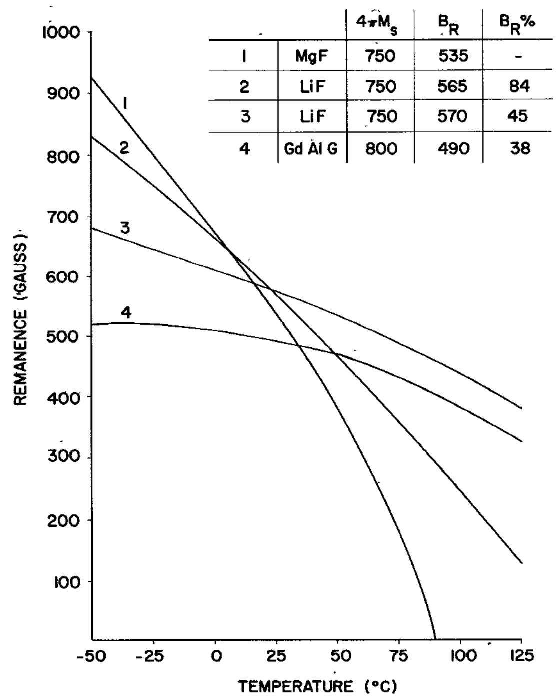

Fig. 7. B_R versus temperature for a magnesium ferrite, lithium ferrites, and a gadolinium-aluminum YIG with optimum $4\pi M_s$ for S-band latching applications.

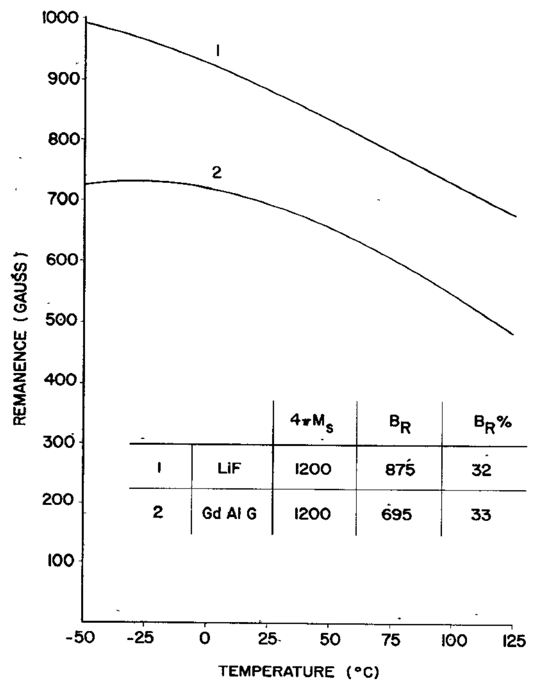

Fig. 8. B_R versus temperature for a lithium ferrite and a gadolinium YIG with optimum $4\pi M_s$ for C-band latching applications.

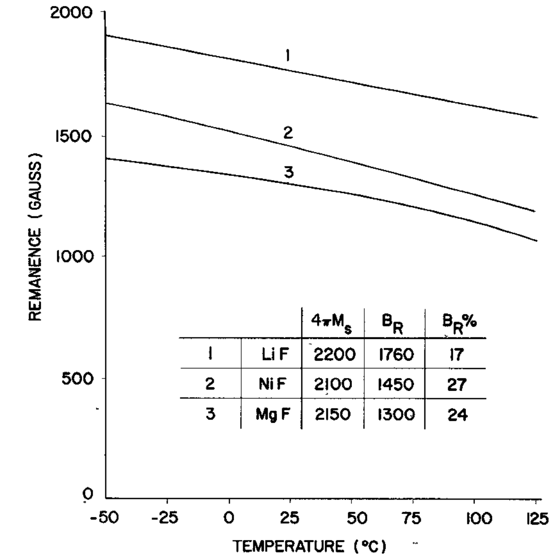

Fig. 9. B_R versus temperature for nickel, magnesium, and lithium ferrites with optimum $4\pi M_s$ for X-band latching applications.

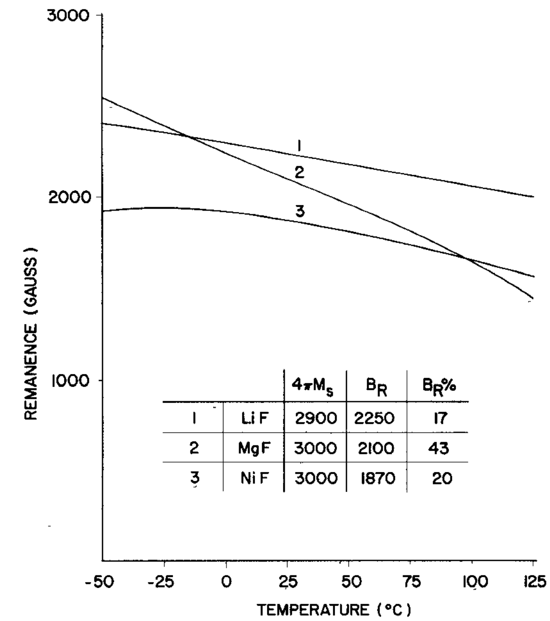

Fig. 10. B_R versus temperature for nickel, magnesium, and lithium ferrites with optimum $4\pi M_s$ for K_u-band latching applications.

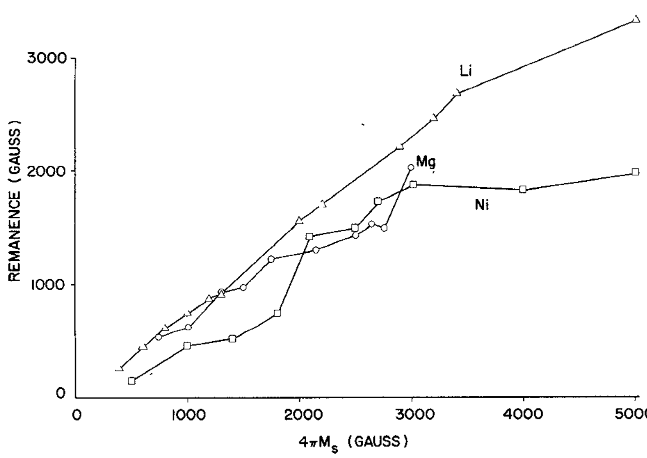

Fig. 11. Remanent magnetization versus saturation magnetization for lithium, magnesium, and nickel ferrites.

D. Cost

The cost of raw materials used in the preparation of ferrimagnetic materials has a major influence on the selling price of phase shifter elements intended for use at S and C bands. The raw materials used in the preparation of most spinel ferrites are about an order of magnitude cheaper than for most members of the Gd YIG system. Reasonable manufacturing yields, raw materials cost, and labor input dictate that a spinel-ferrite S-band element will sell for about 50 percent of the cost of a garnet element. Because the typical C-band configuration is smaller, the raw material content is a lower proportion of the unit cost. Therefore, a spinel C-band element sells for 70 percent of the cost of a garnet element. The difference in price gets proportionately smaller as the frequency of operation increases. Structures for higher frequency applications utilize very little material and the effect of raw material costs essentially drops out of pricing considerations. At such frequencies, the superior performance of lithium ferrites relative to other systems, rather than cost, becomes the deciding factor.

E. Summary

The lithium-ferrite system is comprised of discrete compositions with saturation magnetizations of from < 200 to > 5000 G. Each of these compositions can be varied to optimize the magnetic loss-temperature stability tradeoff via anisotropy reduction for any practical level of power handling capability.

Low magnetization (< 800 G) lithium-titanium ferrites with the best possible temperature performance have inherently high anisotropy, which is responsible for excessive low field magnetic loss. The presently employed method used for reducing anisotropy in the lithium-ferrite system compromises temperature performance but does result in low loss. For a fixed magnetization, power handling capability, and loss the Gd YIG's are less temperature sensitive than low anisotropy lithium-titanium ferrites.

For C-band latching devices, where materials with magnetizations of $\sim$1200 G are normally employed, the level of anisotropy encountered in the best temperature performance lithium ferrites does not influence insertion loss. In this case for garnets to match the temperature stability of the lithium ferrites, they must be heavily doped with Gd^{+3}, and as a result of this are slightly lossier.

Lithium ferrites are superior to the nickel ferrites and magnesium ferrites used at frequencies in the X band and above. Improvements on the order of 15–50 percent in remanence ratio are obtainable with the lithium-ferrite system in comparison with other spinels.

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Diode Phase Shifters for Array Antennas

JOSEPH F. WHITE, MEMBER, IEEE

(Invited Paper)

***Abstract*—This paper contains considerations for diode phase shifters used for phased array antenna control. The categories are: 1) areas in which ferrite and diode phase shifters differ, 2) diode phase-shifter circuits, 3) the nature and typical performance of p-i-n diodes, 4) the requirements of a driver and a typical circuit, and 5) measured performance of phase shifters in *L*, *S*, *C*, and *X* bands.**

I. INTRODUCTION

A. Diodes and Ferrites as Alternatives

THE TWO principal means of providing electronic control of the phase of microwave signals are realized by the diode and the ferrite phase shifters. Both of these circuit approaches have received continuous and enormous developmental effort [1] since about 1960 when the major interest in the electronically controlled phased array antennas began. It is significant that neither technology has totally bettered the other (in the *S*–*X* frequency bands) in more than a decade of intensive investigation which they have received. Furthermore, it is difficult to imagine two more widely dissimilar technical approaches to the same problem.

In principle, it is possible to have a complete understanding of either approach without any familiarity with the other. In practice however, although some array antennas could be designed about either a diode or a ferrite phase shifter, there cannot be a pair of diode and ferrite phase shifters which have identical behavior. These two approaches share a common objective, namely, the steady-state control of the relative phase between input and output ports. However, even in this respect, the one way phase as a function of frequency characteristic is generally different for the two. Certainly, each approach has its own unique character insofar as further requirements—including power handling capability, reciprocity, switch-

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The author is with Microwave Associates, Inc., Burlington, Mass. 01803.