MICROWAVE PROPERTIES OF ARC PLASMA SPRAYED LITHIUM FERRITE *

R. E. KAELBERER, G. O. WHITE and C. E. PATTON

Department of Physics Colorado State University, Fort Collins, Colorado, 80523, U. S. A.

Résumé. — On a obtenu des données micro-ondes fondamentales en bande X sur la largeur de raie effective (ΔH_{eff}) et la largeur de raie des ondes de spin en pompage parallèle (ΔH_k) pour une série de ferrites polycristallins de lithium (aimantation 1 200 G) destinés à des déphaseurs et fabriqués par pulvérisation en plasma d'arc (matériaux APS). A titre de comparaison les mêmes mesures ont été aussi effectuées sur un ferrite de lithium préparé de façon classique. Aux champs faibles et élevés, en dehors du manifold d'ondes de spin, tous les matériaux présentent une largeur de raie effective comparable inférieure à 10 Oe. A l'intérieur du manifold les matériaux APS avec des additions de Zn ont un pic de ΔH_{eff} plus faible que ceux sans addition ; toutefois le pic de largeur de raie effective pour tous les matériaux APS est plus large que celui du matériau classique. Les additions de Zn ont un effet opposé sur les résultats à forte puissance ; ΔH_k est plus large pour les matériaux contenant du Zn.

Abstract. — Fundamental microwave data on the X-band effective linewidth (ΔH_{eff}) and parallel pump spin-wave linewidth (ΔH_k) have been obtained for a series of arc plasma sprayed (APS) polycrystalline, 1 200 G, lithium ferrite phase shifter materials. For comparison, these measurements were also performed for a conventionally sintered lithium ferrite material. At low and high fields, outside the manifold region, all materials had comparable $\Delta H_{eff} < 10$ Oe. Within the manifold region, APS materials with Zn additions had a lower peak ΔH_{eff} than those without, although the peak effective linewidths for all the APS materials were larger than the value measured for the conventional material. Zinc additions had an opposite effect on the high power data where ΔH_k was larger for APS materials with Zn additions.

1. **Introduction.** — Recent interest in the use of arc plasma sprayed (APS) lithium ferrite materials for phased array radar applications [1] has shown the need for a study of the basic microwave properties of these APS materials. The APS process is an economical and promising method for the preparation of ferrite materials for device applications. Two important considerations are : (1) the insertion loss of the device, (2) the microwave power handling capability of the material.

Prototype phase shifters have been produced and evaluated in device configurations [1]. However, very little basic microwave data have been reported for these materials. The present paper reports on measurements of (1) effective linewidth (ΔH_{eff}) *versus* static field at 10 GHz and (2) spin-wave linewidth (ΔH_k) *versus* field over the parallel pumping region (0-1.7 kOe) at 9.2 GHz. These basic data represent the first such comprehensive measurements on arc-plasma sprayed lithium ferrite materials.

The effective linewidth concept was introduced to provide a meaningful loss parameter for ferrite materials biased at fields outside of the resonance region [2]. Phase shifters generally operate at remanence magnetization. Thus, the low field ΔH_{eff} is the relevant loss parameter. The region around resonance (manifold region) is usually characterized by a large increase in ΔH_{eff} due to anisotropy and/or porosity related inhomogeneous broadening or two-magnon scattering [3]. The field dependence of ΔH_{eff} in this region is related to the ferrite microstructure [4]. At high field ΔH_{eff} is generally taken as a measure of the intrinsic losses in the absence of any two-magnon scattering or inhomogeneous broadening. Room temperature results at 9.9 GHz are reported here for all three field regions.

The spin-wave linewidth is the relaxation rate of spin-wave modes, expressed in linewidth units, which are parametrically pumped when the microwave field amplitude (h) exceeds some critical value h_{crit}. For $h > h_{crit}$, the material losses and the related insertion loss increase substantially over the values below h_{crit}. Therefore, h_{crit} is a quantitative measure of the power handling capability of the material. Both h_{crit} and the derived spin-wave linewidth, ΔH_k, exhibit a characteristic field dependence. Results on h_{crit} *versus* static field, the so-called *butterfly curve* for parallel pumping, have been obtained at room temperature and 8.9 GHz. These data were used to calculate the wave number dependence of ΔH_k. This dependence is related to the ferrite microstructure and to grain size effects in particular.

2. **Ferrite materials.** — The arc plasma spray (APS) materials were fabricated and supplied by R. W. Babbitt and coworkers [5]. They consisted of long, rectangular ferrite toroids sprayed directly on dielectric

* Work supported by ARO Contract # CO4-75-6-0010.

cores. The ferrite material was machined to a thickness of about 1.5 mm. To obtain spherical samples for the basic microwave measurements, cubes of ferrite material were cut from the toroids and ground into spheres of 1.0-1.4 mm diameter by a two-pipe lapidary method. The toroids were initially sprayed from two types of powders, Ampex 1202 powder ($Li_{0.79}Fe_{1.63}Ti_{0.57}Mn_{0.03}O_4$) and Ampex 1200 powder (composition similar to 1202 but with Zn additions). Both powders have saturation magnetizations ($4\pi M_s$) on the order of 1200 G. For comparison, samples were also cut and ground from a conventionally sintered 1200 G substituted lithium ferrite material, Trans-Tech L-27-J [6].

Table I lists the samples, powder preparation, grain size, and measured $4\pi M_s$ for the materials studied.

TABLE I

Sample composition and microstructure for the lithium ferrite materials of this study

Sample code number	Sample preparation	Average grain size (μ)	$4\pi M_s$ (G)
APS C-L-113	Ampex 1202 powder fully reacted	7.4	1 240
APS C-L-122	Ampex 1202 powder fully reacted	4.7	1 200
APS C-L-163	Ampex 1202 powder spray dried	11.1	1 140
APS C-L-130	Ampex 1202 powder fully reacted (no anneal)	—	1 210
APS C-L-012	Ampex 1200 powder partially reacted (with Zn addition)	—	1 110
APS C-L-077	Ampex 1200 powder spray dried (with Zn addition)	7.3	1 220
Trans-Tech L-27-J	Conventional firing	large	1 240

The values of $4\pi M_s$ were determined at 8 kOe applied field by standard vibrating sample magnetometry. The $4\pi M_s$ values are accurate to $\pm$ 4 %. The small error is due to an uncertainty in the sample volume determinations on the small spheres. The average grain sizes were estimated from electron microscopy photographs [7].

3. **Effective linewidth.** — The experimental technique used to measure the effective linewidth is an improved version of that reported by Patton and Kohane [8]. The sample is placed in a transmission cavity and the microwave susceptibility is measured as a function of static external magnetic field. The changes in cavity Q and resonant frequency yield the complex microwave susceptibility of the sample. Improvements in sensitivity were achieved by using a high stability counter [9] to directly measure the klystron frequency to within 500 Hz at 10 GHz. The frequency drift of the cavity due to temperature changes around room temperature ambient was reduced when necessary with a temperature stabilized water jacket around the cavity. The cavity temperature could be maintained constant within $\pm$ 0.04 °C at 8 °C.

To measure the effective linewidth in the manifold region [10], from about 3 kOe to 4.5 kOe, a TE_{102} rectangular cavity with a relatively low Q of about 1700 was used. Data were taken at 9.9 GHz and room temperature. Temperature stabilization of the cavity was not necessary. Representative data are shown in figure 1. The field dependence of ΔH_{eff} for all the

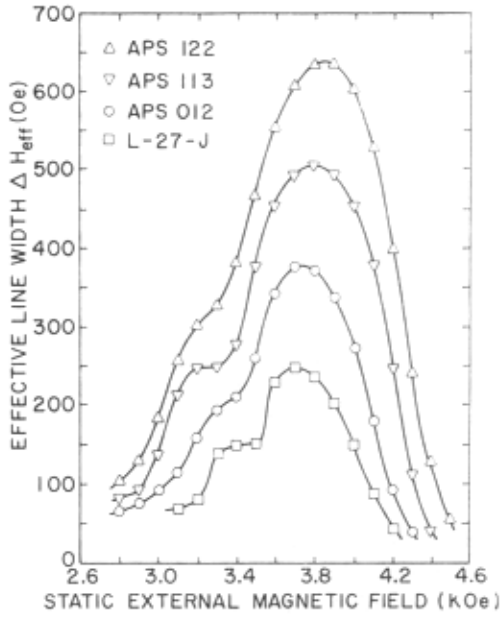

FIG. 1. — Effective line-width as a function of static external magnetic field in the manifold region for the lithium ferrite materials.

samples is similar to that for dense Ca-V substituted YIG with large magnetocrystalline anisotropy and porous YIG with relatively high porosities (> 5 %) [4]. The APS materials exhibit larger losses than the conventional fired L-27-J, presumably an indication of the increased porosity of materials fabricated by the APS method.

Some correlation with composition and grain size can also be seen in the tabulation of the peak ΔH_{eff} values given in table II. The samples are listed in order of decreasing ΔH_{eff}. The four samples with the largest ΔH_{eff} are from the Ampex 1202 powder with no Zn additions. Of this group, the largest value of ΔH_{eff} was measured for the C-L-122 sample which has the smallest average grain size. The two samples, C-L-012 and C-L-077, sprayed from Ampex 1200 powder with Zn additions, have lower peak ΔH_{eff} values. This may be related to a reduction in crystalline anisotropy reported for conventional Zn substituted Li ferrite [11]. The smallest ΔH_{eff} was found for the conventional, commercially prepared Trans-Tech L-

TABLE II

Peak effective linewidth measured in the manifold region, sample composition and average grain size for the lithium ferrite materials.

Sample	Peak H_{eff} (Oe)	Material	Average grain size (μ)
C-L-122	630	APS 1202	4.7
C-L-163	520	APS 1202	11.1
C-L-130	510	APS 1202	—
C-L-113	500	APS 1202	7.4
C-L-012	370	APS 1200 (Zn addition)	—
C-L-077	330	APS 1200 (Zn addition)	7.3
L-27-J	250	Conventional	large

27-J. Although none of the APS materials have effective linewidths as low as the conventionally prepared material, those with Zn additions do appear to have reduced effective linewidths in the manifold region. Although the data are somewhat limited and not completely consistent, it also appears that a small ΔH_{eff} in the manifold region is related to large average grain size.

Outside the manifold region, at fields below 3 kOe or above 4.5 kOe, two-magnon scattering and inhomogeneous broadening related to low k spin-waves are absent [3]. The relaxation rate is considerably reduced, compared to the values in the manifold region. Very accurate measurements of cavity parameters are necessary to determine the effective linewidth. For these off-resonance ΔH_{eff} determinations, a TE_{011} cylindrical cavity with a high Q (20, 500) was used. Measurements were made at 9.8 GHz and the cavity temperature stabilized at 8 °C ± 0.04 °C.

Representative data outside the manifold region are shown in figure 2. These ΔH_{eff} values are smaller than the values measured in the manifold region (Fig. 1) by a factor of 50. For these off-resonance ΔH_{eff} data, there appears to be no consistent correlation between APS parameters and the losses. The effective linewidths at high field for the APS materials are about 5-10 Oe. The conventionally prepared L-27-J has a high field ΔH_{eff} of about 4 Oe. These values are quite comparable. For fields around 2 kOe, the ΔH_{eff} for all samples is slightly higher than at high field, on the order of 10 Oe. Precise measurements are precluded by the large inhomogeneous broadening contributions which expand the manifold region, and by experimental problems at low fields (< 2 kOe) due to low susceptibilities and small samples.

It is clear, however, that the relevant linewidth parameter for the APS materials in phase shifter applications is on the order of 5-10 Oe. This value is comparable to the low-field ΔH_{eff} for conventionally prepared materials.

4. **Spin-wave linewidth.** — Spin-wave thresholds (h_{crit}) and linewidths (ΔH_k) for the various lithium ferrite materials were determined from the microwave field dependence of the parallel pump susceptibility. The signal source was a 2J51 tunable X-band magnetron driven by a modified MIT Model 3 hard tube modulator [12]. Microwave pulses 3 μs long at 8.96 GHz, with a 200 Hz repetition rate were used in conjunction with a rectangular TE_{101} reflection cavity. Standard microwave techniques were used to determine the negative, imaginary part of the sample susceptibility (χ'') as a function of microwave field amplitude h. The threshold field h_{crit} was estimated from graphs of χ'' *versus* h.

The microwave threshold field (h_{crit}) for parallel pump spin-wave instability is shown as a function of static applied field in figure 3. Here samples C-L-122

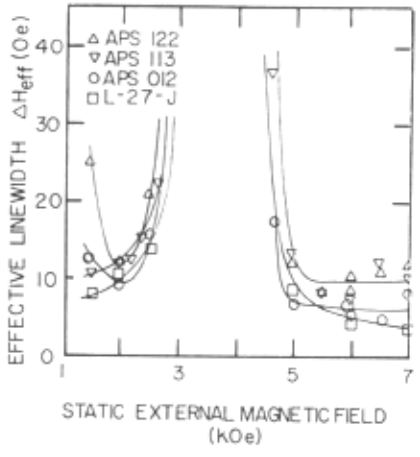

FIG. 2. — Effective linewidth as a function of static external magnetic field outside the manifold region for the lithium ferrite materials.

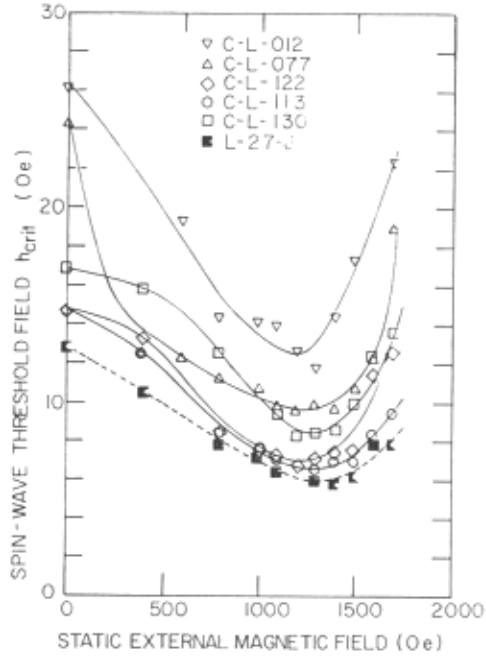

FIG. 3. — Spin-wave threshold field as a function of static external magnetic field for the lithium ferrite materials.

and C-L-113, sprayed from Ampex 1202 powder and annealed, exhibit values of h_{crit} only slightly higher than those for the commercial Trans-Tech L-27-J material. The C-L-130 sample, sprayed from 1202 powder but not annealed, has significantly higher values of h_{crit}. The C-L-012 and C-L-077 samples were sprayed from Ampex 1200 powder which contains a small amount of Zn (approximately 0.05 Zn atoms per formula unit). It is to be noted that the magnetization of sample C-L-012 may be quite non-uniform [13]. This non-uniform $4\pi M_s$ is a likely cause of the large microwave threshold field in this sample, compared to the C-L-077. It is clear, however, that both 1200 powder samples exhibit higher thresholds than those of the 1202 powder samples. The following conclusions are suggested by the data of figure 3. (1) The results for the 1202 powder samples (C-L-113, C-L-122, C-L-130) indicate that the APS process can produce a significant increase in h_{crit}, most of which is lost upon annealing [1]. (2) Comparison of the 1200 powder samples (C-L-012, C-L-077) with the 1202 powder samples (C-L-113, C-L-122, C-L-130) shows that Zn additions increase the microwave power capability for APS materials, as has been noted for conventionaly sintered ferrites [11]. (3) An inhomogeneous magnetization, which can be produced by the APS process as for C-L-012, may be reflected in large apparent h_{crit} values.

The wave number dependence of the spin-wave linewidth (ΔH_k) may be obtained from the Suhl-Schlömann spin-wave instability theory [14, 15] and the butterfly curve data of figure 3 [16]. These results are shown in figure 4. The sample to sample

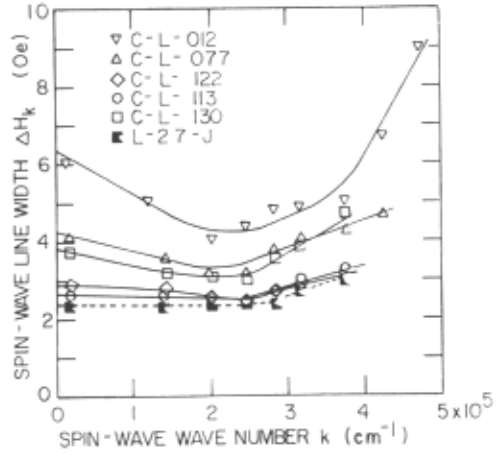

FIG. 4. — Spin-wave line-width as a function of spin-wave numbers for the lithium ferrite materials.

variation in ΔH_k is qualitatively similar to the variation in h_{crit}, with $\Delta H_k(k \to 0)$ values ranging from 2.6 Oe for the conventionally sintered L-27-J, to 6 Oe for the Zn doped APS C-L-012.

Of fundamental interest is the ΔH_k *versus* k behaviour summarized in figure 4. Below $k \approx 2 \times 10^5$ cm^{-1}, ΔH_k is a decreasing function of k. Above $k \approx 2 \times 10^5$ cm^{-1}, ΔH_k is an increasing function of k. This general behaviour is qualitatively similar to the behaviour for fine grain and porous YIG [17, 18]. In YIG, the inverse k-dependence of ΔH_k below $k \approx 2 \times 10^5$ cm^{-1} has been qualitatively related to spin-wave transit time arguments [17, 18]. The available microstructure data on the APS materials also indicate relatively small grains. Therefore, it is likely that the ΔH_k *versus* k dependence in figure 4 for the APS materials is related to grain size effects.

5. **Discussion.** — The large values of ΔH_{eff}, up to 650 Oe, in the manifold region are clearly related to the porosity and microstructure inherent in the APS process. The values are reduced by the addition of Zn. At low field ($\sim$ 2 Oe) or above 5 kOe, the effective linewidths of the APS materials are less than 10 Oe, comparable to those of conventionally prepared materials. The variations observed from sample to sample show no clear microstructure correlations. The high power data show that Zn additions increase h_{crit} and ΔH_k, opposite to the effect of Zn on ΔH_{eff} in the manifold region. This result is quite peculiar. The ΔH_{eff} in the manifold region and the high h_{crit} values must both be microstructure related. It is perplexing that a microstructure effect, which increases ΔH_{eff}, can cause a decrease in ΔH_k. Some clues to this behaviour may be inferred from previous work on the effect of Zn. As previously mentioned, Baba, *et al.* [11] report that Zn reduces the magnetocrystalline anisotropy. This may be the cause of the reduction in ΔH_{eff}. West and Blankenship [19] report that Zn can increase porosity in hot pressed Li-ferrite. This may be related to the ΔH_k increase.

The most important practical observations are : (1) zinc substitution increases ΔH_k, while decreasing ΔH_{eff} in the manifold region ; (2) the off-resonance loss properties are comparable to the losses in conventional materials ; (3) the high power capability is the same or better than that of conventional materials. These observations in combination with the economic advantages of the APS process indicate the utility of these lithium ferrite materials for phase shifter applications.

References

[1] BABBITT, R. W., *IEEE Trans. Mag.* **MAG-11** (1975) 1253.
[2] KOHANE, T. and SCHLÖMANN, E., *J. Appl. Phys.* **39** (1968) 720.
[3] PATTON C. E., *IEEE Trans. Mag.* **MAG-8** (1972) 433.
[4] PATTON, C. E., *Phys. Rev.* **179** (1969) 352.
[5] APS materials supplied by R. W. Babbitt, Army Electronic Command, Fort Monmouth, New Jersey, U. S. A.
[6] Trans-Tech L-27-J lithium ferrite supplied by R. West, Trans-Tech, Inc., Gaithersburg, Maryland, U. S. A.

[7] Data on microstructure provided by A. Tauber, Army Electronic Command, Fort Monmouth, New Jersey, U. S. A.
[8] PATTON, C. E. and KOHANE, T., *Rev. Sci. Instr.* **43** (1972) 76.
[9] Model 5340A Hewlett Packard frequency counter.
[10] The manifold region is defined as the field region in which the uniform precession is degenerate with low wave number spin-wave states.
[11] BABA, P. D., ARGENTINA, G. M., COURTNEY, W. E., DIONNE, G. F., and TEMME, D. H., *IEEE Trans. Mag.* **MAG-8** (1972) 83.
[12] The MIT Model 3 pulser, which is capable of delivering 144 KW pulses to the magnetron, is inexpensively available surplus for $50-$300. Its modification will be discussed in a future publication.
[13] TAUBER, A., private communication.
[14] SUHL, H., *J. Phys. Chem. Solids* **1** (1957) 209.
[15] SCHLÖMANN, E., Raytheon technical Report R-48, October 1959.
[16] The use of data on h_{crit} *versus* H to calculate ΔH_k *versus* wavenumber k requires a numerical value for the exchange parameter D. In order to obtain ΔH_k *versus* k, the YIG value (see Ref. [18]) $D = 5 \times 10^9$ Oe cm^2 has been used. This is a reasonable approximation because the Curie temperatures are the same to within about 15 %. Any error in k, however, is probably less than this, since k varies as $\sqrt{D}$.
[17] PATTON, C. E., *J. Appl. Phys.* **41** (1970) 1637.
[18] PATTON, C. E., *Proc. Intl. Conf. on Ferrites, Kyoto*, Japan (U. of Tokyo Press, Tokyo) (1970) p. 524.
[19] WEST, R. G., and BLANKENSHIP, A. C., *J. Amer. Ceram. Soc.* **50** (1967) 343.