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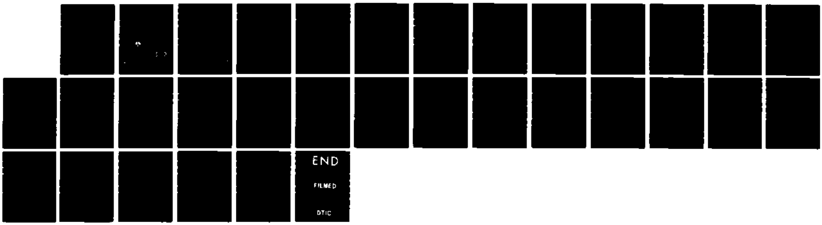

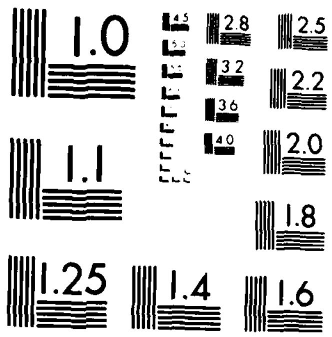
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FINAL TECHNICAL REPORT

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By

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The objective of this research is to develop new and improved epitaxial ferrite materials for use in microwave and millimeter-wave signal processing devices. The major emphasis has been on multiple layer magnetic garnet structures for magnetostatic wave (MSW) delay lines. Research performed under a previous contract demonstrated that improved linearly dispersive MSW characteristics (that is, linear variation of delay time with frequency) could be obtained using structures which consisted of two epitaxial magnetic garnet layers

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SECURITY CLASSIFICATION OF THIS PAGE (When Data Entered) separated by an epitaxial nonmagnetic layer. More detailed analysis of the magnetostatic modes in such multiple layer materials has been carried out under the present contract using ferromagnetic resonance (FMR) spectroscopy. This work is aimed at understanding details such as the occurrence of notches in the passband of multiple layer MSW delay lines. A significant problem, which is common to all MSW delay lines, single layer as well as multiple layer, is the presence of fluctuations in the delay vs. frequency characteristics. These fluctuations, usually called ripple, are attributed to reflections of the propagating magnetostatic waves. A new method for suppressing ripple has been devised and demonstrated. In addition to the work on magnetic garnets, research on epitaxial growth of other ferrites has been continued. In the course of studying epitaxial growth of strontium hexaferrites, a new ferrite material was developed. Some research on the conditions for preparing this new material was carried out. Crystal growth of gallate spinels for use as substrates for epitaxial growth of lithium ferrite and hexagonal ferrite was continued with further improvements in substrate size. Work on sputter deposition of hexagonal ferrites was initiated.

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EPITAXIAL GARNETS AND HEXAGONAL FERRITES

AFOSR Contract F49620-82-C-0081

82 JUN 15 to 83 JUN 14

Final Technical Report

H. L. Glass and L. R. Adkins, Principal Investigators

December 1, 1983

I. OBJECTIVES

The overall objective of this research is to develop epitaxial single crystal ferrite films suitable for microwave and millimeter-wave signal processing at frequencies above 1 GHz. The specific tasks are:

1. Analyze and develop special layered garnet structures for propagating-magnetic-wave devices operating at frequencies between 1 GHz and 25 GHz.

2. Investigate LPE growth of lithium ferrite with the objective of preparing low-loss, large area films suitable for delay lines.

3. Investigate epitaxial hexagonal ferrites for use in devices operating at frequencies above 70 GHz, including investigation of substitutions such as Ga or Al as a method for raising anisotropy fields to produce materials useful for devices operating at frequencies above 100 GHz.

These tasks represent a continuation of the work done under previous contracts F44620-75-C-0045, F49620-79-C-0048, and F49620-80-C-0045. Task 1 relates to magnetostatic wave (MSW) devices similar to surface accustic wave (SAW) devices, but operating at higher frequencies. This task makes use of the fairly well established epitaxial garnet technology. Tasks 2 and 3 represent an exploration of the epitaxial growth of other classes of ferrites which have capabilities beyond those of the garnet-structure ferrites.

II. BACKGROUND

In order to appreciate the accomplishments under this contract, it is perhaps worthwhile to consider the status of epitaxial garnets and hexagonal ferrites when the series of contracts began in 1975. At that time the liquid phase epitaxy method for growth of magnetic garnet films was quite new. It appeared to be capable of producing films which were as crystallographically perfect as the substrates. The substrates, rare earth gallium garnets, especially gadolinium gallium garnet (GGG), were becoming commercially available and had extremely low defect densities. The principal interest in epitaxial garnets was for magnetic bubble memories. For this application the films had to be about 3μm thick with low defect density and excellent thickness and compositional uniformity. Low microwave losses were not a requirement. In fact, rather substantial magnetic damping was desirable as a means of maintaining stable domain wall structures.

The major initial thrust of this series of contracts was to optimize the liquid phase epitaxy (LPE) method for growth of microwave garnets, specifically yttrium iron garnet (YIG). The major attraction of YIG was its low microwave losses, corresponding to narrow ferromagnetic resonance peaks. The theoretical value for the intrinsic linewidth was 0.2 Oe at X-band (10 GHz). Values close to this had been reported for spheres fabricated from flux-grown bulk crystals. Single crystal films of YIG were potentially attractive for applications where spheres or bulk crystals were unsuitable (such as magnetostatic wave devices); for device designs compatible with planar waveguide and microwave integrated circuits; or perhaps as a lower cost alternative to spheres. However, previous attempts to prepare single crystal films of YIG had not been successful in attaining low losses. Also, previous attempts had failed to yield good quality films of the thickness needed for microwave devices.

During the early years of this research, articles published by other workers indicated that the LPE process had inherent characteristics which would prevent successful growth of thick films and which would limit the attainable linewidth. The difficulties with thick films were not merely that misfit stresses would cause fracture but that the LPE process entailed an instability at the crystal growth interface which would cause progressive development of hillocks and ridges. Thus, there appeared to be a limit of about 30μm beyond which crystal quality deteriorated. The difficulty in attaining low linewidths was the substrate, which contained magnetic rare earth impurities that would create direct relaxation loss processes and gallium that would produce gradients in the magnetization. Even if these could be controlled, incorporation of Pb, an essential component in the LPE solvent, would create valence exchange loss mechanisms.

Probably the most significant accomplishments during the early years of the contracts were the attainment of intrinsic losses (0.15 Oe linewidth at 9.5 GHz) and the demonstration of high quality films 100μm thick. The key to attaining low losses was to avoid contamination by magnetic impurities and to deliberately incorporate Pb to charge compensate other impurities and point defects. The ability to grow high quality thick films was based in part on using the incorporated Pb to reduce misfit stress but, more importantly, using well polished substrates and clean deposition conditions to prevent nucleation of ridges and hillocks. At the present time we grow low loss, 50μm thick films routinely for use in magnetostatic wave devices for two current Air Force contracts. In addition, over the years of this series of contracts, we have supplied films to several universities and a number of government laboratories.

Once the techniques for growing low linewidth YIG films of suitable thickness were established, attention was focused on two problems which were important to the successful application of epitaxial YIG in microwave devices. One problem was the variation of magnetic and microwave properties with temperature. Of course in practice this problem could be solved by external methods such as temperature control or electronic compensation. However, we devoted some effort to understanding the nature of magnetic anisotropy in epitaxial garnets. This led to the demonstration of modified YIG compositions in which the various anisotropy contributions could be counterbalanced to adjust the temperature variation and even to achieve a ferromagnetic resonance condition which was constant over a significant range of temperatures.

Temperature stability was achieved by using gallium as a substitute for part of the iron and lanthanum as a substitute for part of the yttrium in the YIG films. This experience was of great use in solving the second problem. This problem was to prepare a material which was optimized for use in magnetostatic wave devices. Such devices are in many ways analogous to surface acoustic wave devices. Typically they take the form of a delay line. An input transducer launches a wave (a magnetic spin wave in the magnetostatic wave device, an acoustic wave in the surface acoustic wave device) which propagates along the crystal and is picked up by an output transducer. While surface acoustic wave devices are useful up to about 1 GHz, magnetostatic wave devices are useful from about 1 GHz up through the microwave frequency range. Also, magnetostatic wave devices are tunable; that is, the center frequency can be adjusted by choosing the strength of an externally applied magnetic field.

Low linewidth is important for magnetostatic wave devices where it corresponds to low propagation loss. Just as important, however, is control of dispersion, the variation of delay time with frequency. For most applications, nondispersive (delay time is constant for all frequencies in the operating bandwidth) or linearly dispersive (delay time is proportional to frequency) behavior is required. In general, magnetostatic wave delay lines exhibit complicated nonlinear dispersion.

A very promising method for dispersion control is to use multiple layer YIG instead of a uniform single layer. The dispersion can be adjusted by choice of the number of layers and the thickness, saturation magnetization and magnetic anisotropy of each. To accomplish this in practice requires a good theoretical model of magnetostatic wave propagation in multiple layer structures and the ability to prepare these structures while retaining high crystal quality and low losses. A major area of activity during the past few years of the research contracts has been to develop models for the three principal modes of magnetostatic wave propagation: magnetostatic surface waves (MSSW), magnetostatic forward volume waves (MSFVW), magnetostatic backward volume waves (MSBVW). The models are used to determine the optimum multiple layer structures for nondispersive or linearly dispersive delay characteristics.

Multiple layer structures are grown by successive liquid phase epitaxy. Lanthanum and gallium (or aluminum) substitutions are used to alter the magnetic properties of the layers. The level of substitution can be sufficient to reduce the saturation magnetization to such an extent that the layer behaves as though it were a nonmagnetic dielectic. Thus magnetic and monmagnetic layers can be grown having thicknesses ranging from about 1μm to about 100μm.

Although the LPE process is highly reproducible and the effects of lanthanum and gallium substitution are fairly well understood, the quantitative relationship between LPE growth conditions and the magnetic properties of the layers must be determined empirically. This is best done by ferromagnetic resonance (FMR). The FMR spectra of multiple layer materials are quite complicated. Therefore, part of the research effort has been directed toward a theoretical analysis of FMR in multiple layer materials.

Multiple layer structures may seem complicated. The fact is, however, that once the required structure has been defined it is fairly easy to obtain by LPE. In the course of this research we have demonstrated nondispersive delay lines with flatter characteristics than any previously reported. In addition we have demonstrated linearly dispersive delay lines having linearity at least as good as any previously reported. Moreover, our multiple layer linearly dispersive delay lines have the potential for significantly lower propagation loss than alternative designs.

A continuing problem with magnetostatic wave delay lines is ripple. Ripple, observed as fluctuations in the delay time versus frequency characteristics, is attributable to reflections of the propagating magnetostatic waves. Various methods have been tried by different investigators to prevent or attenuate reflections. Methods include beveling the YIG layers, damaging the end regions of the delay lines, distorting the applied magnetic bias field near the ends of the delay lines. In recent work on this contract we have demonstrated a new method of ripple suppresion which employs an epitaxial attenuation layer. Thus, the new method is an extension of our work on multiple layer YIG.

At the present time, YIG is the only ferrite which can be grown in the form of large-area layers having low linewidth. Other potentially low linewidth ferrites are also of interest; especially lithium ferrite and hexagonal ferrites. Lithium ferrite is of interest mainly because it has a higher saturation magnetization than YIG. This property can result in larger bandwidths and higher operating frequencies in devices. In addition, lithium ferrite has a higher Curie temperature which may make it easier to obtain temperature stable device performance. Hexagonal ferrites are of interest mainly because of their high magnetocrystalline anisotropy. This can function like a built-in magnetic field to bias resonance to high frequencies. Thus, hexagonal ferrites can be used in devices which operate at millimeter-wave frequencies using magnets which would provide only microwave frequency operation for other ferrities.

LPE growth of YIG was facilitated by the existence of a suitable substrate, GGG, which was commercially available. For epitaxial growth of lithium ferrite and hexagonal ferrites, suitable substrates have had to be identified, grown, and evaluated. During the earliest period of the series of contracts we used bulk crystals of hexagonal ferrites as substrates and were able to demonstrate the first LPE growth of hexagonal ferrite. Subsequently, we grew crystals of zinc gallate spinel and magnesium indium-gallate spinel and, using these, were the first to report LPE growth of hexagonal ferrites (several different compositions and types) on non-magnetic substrates. In these cases, (111) surfaces of the spinel crystals were used because of the structural similarity to the basal planes of hexagonal ferrite. We also used these substrates for LPE growth of lithium ferrite which, like the substrates, has the spinel crystal structure. We were among the first to report LPE growth of lithium ferrite and we have reported the lowest linewidth, less than 8 Oe at 9 GHz.

Investigations of LPE growth of lithium and hexagonal ferrites have continued throughout the series of contracts, although at a lower level of effort than the YIG work. Improvements have been made in the techniques for growing the gallate spinel substrate crystals so that substrates 1 or 2 cm in diameter are obtained fairly reproducibly. Some work has been done on preparing non-(111)-oriented substrates for use in LPE of lithium ferrite.

Various LPE melt compositions were studied to try to improve the quality (espically the uniformity) of lithium ferrite and hexagonal ferrite layers. Also, modifications to the chemical composition of the hexagonal ferrites were investigated to demonstrate the ability to increase the anisotropy fields and, thus, increase the frequency useful for device operation. Eventually, 94 GHz center frequencies should be attained easily.

In summary, when this series of contracts began, LPE growth of YIG on GGG was known; however, there were serious doubts about the ability to prepare YIG layers suitable for microwave devices. Specifically, there were doubts about the ability to obtain very low linewidths and to obtain good quality layers of sufficient thickness. LPE growth of lithium ferrite and hexagonal ferrites had not been reported and suitable substrates had not been identified. The research performed under this series of contracts has b significant in solving the basic problems with LPE YIG so that high qual ty, low linewidth layers of thickness suitable for microwave devices are avai able. In addition, considerable progress was made in advanced LPE YIG, especially multiple layer materials for dispersion control in magnetostatic wave devices. LPE of lithium ferrite and of several hexagonal ferrites was developed using gallate spinel crystals as substrate. Large area layers of very low linewidth have not yet been demonstrated for these ferrites. Further research is required to improve these materials. The research probably should include evaluation of other substrate materials as well as study of ferrite growth processes.

III ACCOMPLISHMENTS

As described in the previous section, a major problem with existing magnetostatic wave (MSW) delay lines is ripple. This is observed in the form of fluctuations in the delay-time versus frequency dispersion curves. It also shows up as fluctuations in attenuation versus frequency across the passband. Ripple is attributable to reflections of the magnetostatic waves, especially reflections from the ends of the delay lines. A new method of ripple suppression was conceived in the course of work we were performing on another Air Force contract (F19628-82-C-0098 with the Electronic Systems

Division of RADC). This contract calls for advanced research on variable nondispersive MSW delay lines. Ripple is a serious problem for the envisioned application. Although conceived on the RADC contract, the method was reduced to practice under this AFOSR contract.

Our method for ripple suppression is a variation of our multiple layer approach to dispersion control. If two adjacent YIG layers have substantially equal magnetic properties, they will behave like a single combined layer as far as magnetostatic wave propagation is concerned; that is, the waves will propagate within both layers. If one of the layers has high propagation losses, then the wave will be attenuated. The basic idea of the method is to provide such as attenuation layer only in regions where high propagation losses are desired; for example, near the ends of the delay line.

To demonstrate this method, we used an ordinary LPE YIG film for one layer (the propagation layer) and an LPE YIG film with a slight holmium substitution for the other layer (the attenuation layer). Holmium is a magnetic rare-earth which has relatively large magnetic moment and orbital angular momentum. Holmium replaces yttrium in the YIG crystal structure. The large magnetic moment provides coupling to the iron spins and, therefore, to the magnetostatic waves. The large orbital angular momentum provides, through spin-orbit coupling, a rather efficient mechanism for transfering energy from the spin system into lattice vibrations. In the attenuation layer, the presence of a few percent holmium substituting for yttrium increases the microwave losses tremendously. The FMR linewidth is 100 Oe or more compared with values much less than 1 Oe for the unsubstituted YIG in the propagation layer. Yet, because the holmium contents is very small, the magnetic properties other than linewidth are substantially unaffected.

Figures (1) and (2) illustrate this. Figure (1) is the FMR spectrum of a non-holmium substituted LPE grown YIG film on GGG. In this case the magnetic bias field is perpendicular to the plane of the film and the applied microwave energy has a frequency of 9.5 GHz. Note that the main resonance line is very narrow (<1 Oe) and this resonance occurs at 4879 Oe.

5. M. T. Elliott and H. L. Glass, "Growth-Induced Anisotropy in Yttrium Iron Garnet Films Grown by Liquid Phase Epitaxy", AIP Conf. Proc. 29 (1976) 115-116.

6. H. L. Glass, "Growth of Thick Single-Crystal Layers of Yttrium Iron Garnet by Liquid Phase Epitaxy", J. Crystal Growth 33 (1976) 183-184.

7. H. L. Glass, M. T. Elliott and F. S. Stearns, "LPE Ferrites for Microwave Devices", American Association for Crystal Growth Western Regional Section, Los Angeles, June 1976.

8. H. L. Glass, M. T. Elliott, "Attainment of the Intrinsic FMR Linewidth in Yttrium Iron Garnet Films Grown by Liquid Phase Epitaxy", J. Crystal Growth 34 (1976) 285-288.

9. F. S. Stearns and H. L. Glass, "Liquid Phase Epitaxy of Hexagonal Ferrites and Spinal Ferrites on Non-Magnetic Spinel Substrates", Mat. Res. Bull. 11 (1976) 1319-1326.

10. H. L. Glass, "Annealing-Induced Relief of Compressive Misfit Strain in Epitaxial Garnet Films", American Crystallographic Association Winter Meeting, Asilomar, California, Feb. 1977.

11. H. L. Glass, J. H. W. Liew and M. T. Elliott, "Temperature Stabilization of Ferrimagnetic Resonance Field in Epitaxial YIG by Ga, La Substitution", Mat. Res. Bull. 12 (1977) 735-740.

12. H. L. Glass, "Annealing-Induced Relief of Compressive Misfit Strain in Liquid Phase Epitaxial Yttrium Iron Garnet Films", J. Crystal Growth 40 (1977) 205-213.

13. H. L. Glass and F. S. Stearns, "Growth of Hexagonal Ferrite Films by Liquid Phase Epitaxy", 1977 INTERMAG Conference IEEE Trans. Mag. MAG-13 (1977) 1241-1243.

Initial sputter deposition experiments employed an aluminum substituted hexagonal ferrite target. The aluminum substitution increases the anisotropy field and, thus, increases the resonance frequency. As the contract year drew to a close, we had succeeded in depositing ferrite. However, the deposit was not epitaxial and the ferrite type could not be identified unambiguously. The work is continuing under new contracts.

IV PERSONNEL

The Principal Investigators were H. L. Glass, who was responsible for crystallographic characterization, and L. R. Adkins, who was responsible for FMR and microwave analyses. F. S. Stearns carried out much of the crystal growth and crystal processing. C. B. Weible assisted with ceramic sample preparation and X-ray analysis. During prior years of the series of contracts, M. T. Elliot and J. H. W. Liaw were major contributors in the area of FMR analysis of the ferrite materials. D. Medellin developed the processes for substrate polishing.

V LIST OF PUBLICATIONS AND CONFERENCE PRESENTATIONS RESULTING FROM AFOSR SUPPORT

1. H. L. Glass and M. T. Elliott, "The Effects of Pb Incorporation on the Ferrimagnetic Resonance Properties of Yttrium Iron Garnet Films Grown by Liquid Phase Epitaxy," Third American Conference on Crystal Growth, Stanford Uniersity, July 1975.

2. H. L. Glass and M. T. Elliott and D. M. Heinz, "Site Preference in Magnetic Garnet Films Grown by Liquid Phase Epitaxy", Tenth International Congress of Crystallography, Amsterdam, August 1975.

3. F. S. Stearns and H. L. Glass, "Liquid Phase Epitaxy of Hexagonal Ferrites", Mat. Res. Bull. 10 (1975) 1255-1258.

4. M. T. Elliott, "Effects of Lead Incorporation of the Ferromagnetic Resonance Linewidths of Liquid Phase Epitaxial Grown Yttrium Iron Garnet", AIP Conf. Proc. 29 (1976) 676-677.

at Fort Monmouth showed that the ceramic meterial was ferromagnetic (or ferrimagnetic) at low temperature. The combination of spontaneous magnetization and transparency to visible light is rather rare. This strontium ferrite borate material may prove to be useful in magneto-optical devices.

A few experiments were performed to determine whether a corresponding compound existed with BaO in place of SrO. No evidence for such a phase was found.

Returning to the subject of hexagonal ferrite, the LPE strontium hexaferrite grown under the Army contract represented a substantial improvement over the prior materials. However, the layers were not smooth. They exhibited the hexagonal scale-like morphology just like the BaO- and PbO-based LPE hexagonal ferrites. On the other hand, work done much earlier in this series of contracts showed that smooth layers could be obtained when the substrates were themselves hexagonal ferrite. Thus, the problem may be ascribed to a chemical mismatch between epitaxial layer and substrate even though there is a good physical (structural) match.

Recently, two research groups in Japan reported success in sputter deposition of barium hexaferrite in amorphous and polycrystalline forms. With an appropriate substrate, perhaps the (111) gallate spinels, it may be possible to obtain a single crystal layer of hexagonal ferrite. This may be an alternative to LPE; or, LPE may be carried out to deposit a hexagonal ferrite layer on top of the sputter deposited layer.

With these possibilities in mind, we began experiments on sputter deposition of hexagonal ferrite. The first step was to modify a sputtering system to accept small targets. This enabled us to make targets ourselves instead of purchasing the standard large targets. This saves time and money while we determine the proper target composition.

year came to a close, there were substrate crystals in various stages of processing as well as a sufficient number of completed substrates to resume the LPE growth experiments. These experiments will be performed as part of the new contract F19628-83-C-0132.

As previously mentioned, the gallate spinel crystals are also used as substrates for epitaxial growth of hexagonal ferrites. In this case, it is the (111) orientation which is of interest because of the structural similarity between the spinel (111) planes and the (00•1) basal planes of the hexagonal ferrites. The most common hexagonal ferrite, barium hexaferrite ($BaO\ 6F2_2O_3$), has an a lattice parameter of 5.892Å which will give a good lattice match if the spinel lattice parameter is 8.33Å. The lattice matching requirements for lead hexaferrite and strontium hexaferrite are nearly the same.

In prior years, we had investigated LPE growth of the BaO- and PbO-based hexagonal ferrites. When the most recent contract year began, we were in the final months of a two year contract with the Army Research Office (DAAG29-80-C-0150) on which we were studying LPE growth of SrO-based hexagonal ferrites. That work, which drew heavily on prior accomplishments under AFOSR support, resulted in epitaxial layers that provided good coverage of the spinel substrates and linewidths as low as 60 Oe at 60 GHz. This represented a substantial improvement over previous LPE hexagonal ferrites which, typically, exhibited linewidths of several hundred Oe at this frequency.

The work on the Army contract also led to the discovery of a new ferrite material which was prepared in single crystal form. The crystals were green, transparent to visible light and exhibited ferromagnetic resonance at temperatures somewhat below room temperature. Chemical analysis showed that these crystals contained boron, as well as strontium and iron, as the major metallic constituents. After conclusion of the Army contract, some experiments were performed on the AFOSR contract to define the composition of this material and to prepare it in ceramic form. The results showed that the composition is $3SrO \bullet Fe_2O_3 \bullet B_2O_3$. Low temperature magnetometer measurements performed at the Electronics Technology and Devices Laboratory

The layer quality was fairly good. Optical microscopy showed that the substrates were covered uniformly and the layer surfaces were smooth. However, growth terraces were present. Using the best growth conditions, a layer was grown on one of the larger substrates, about 1.5 cm in diameter. We tried to measure magnetostatic wave propagation in this sample. However, no output signal was detected. Probably the propagation losses were too high.

The growth terraces observed in these layers are characteristic of growth on facet surfaces. These are the densely packed crystal planes and correspond to the slowest growth directions. For spinel-structured crystals such as lithium ferrite, (111) planes are facet orientations. Not only do the terraces represent physical discontinuities, but they also indicate inhomogeneous growth conditions. To try to circumvent this problem, we decided to prepare substrates of (100) and (110) orientation. These are not facet orientations and may give more uniform growth.

Since the (111) planes are the facet surfaces in spinel, the crystals grow with an octahedral morphology, the octahedral surfaces being the various (111) planes. In order to prepare (100) and (110) substrates from such crystals it is desirable for the crystals to be as large as possible. During the previous contract periods we had investigated methods for obtaining large crystals by modifications to the flux method. One modification was localized cooling of the crucible bottom to restrict nucleation. This tends to give one large crystal rather than many small ones. Other modifications included various types of seeding as an alternative to spontaneous nucleation. In all cases, the flux was PbO - B_2O_3 which, if very high temperatures are avoided, it not excessively volatile. Thus, top-seeding with an open crucible is possible. (Some of our techniques for growing large crystals were described in a presentation to the 6th Conference on Crystal Growth in June, 1982).

Several flux-growth runs of gallate spinels were carried out. Not every run was successful, but quite a few crystals of useful size (1 to 2 cm diameter) were obtained. From these, slices of the desired orientations were cut using a wire saw. The slices were lapped and polished. As the contract

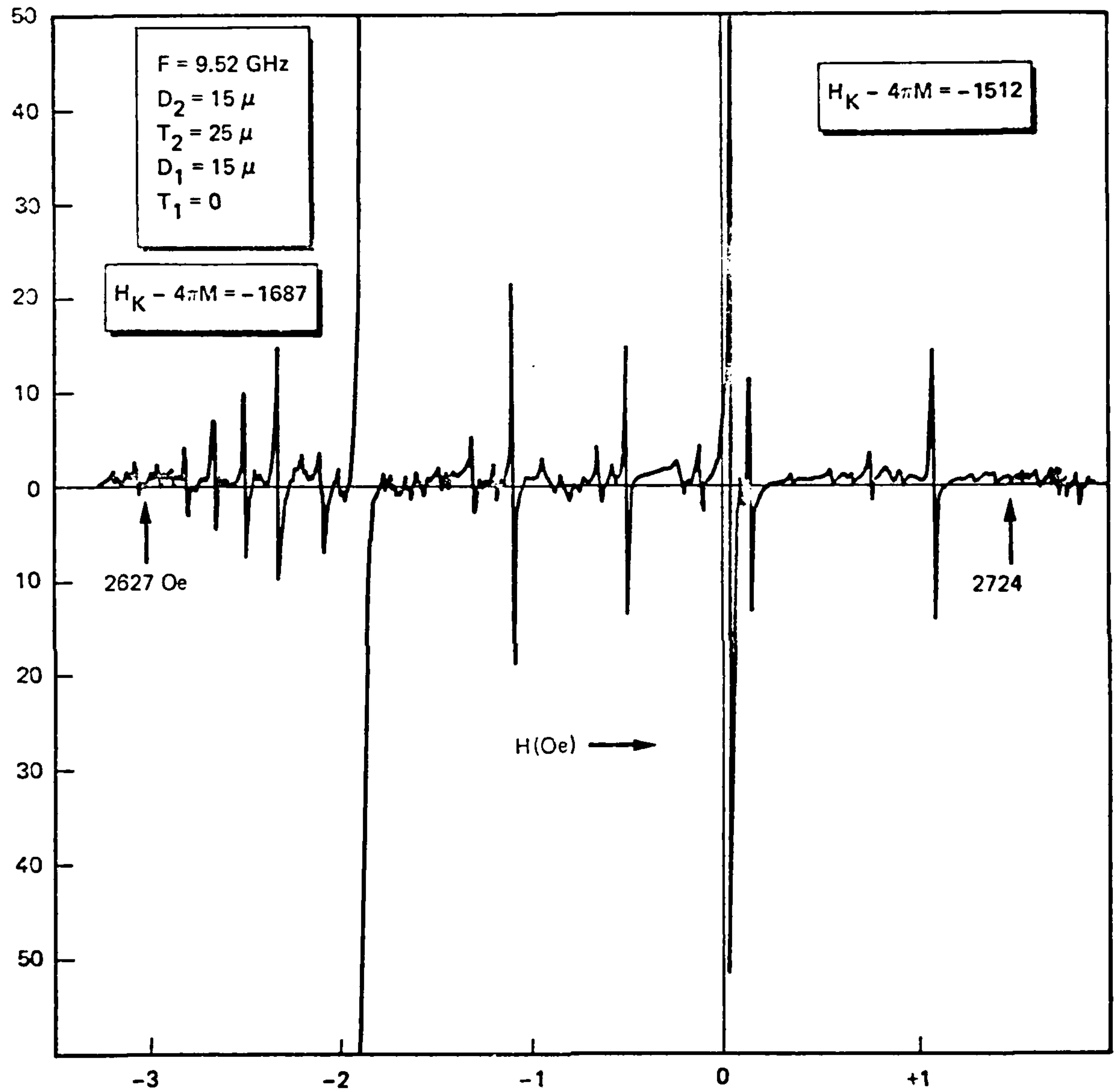

Figure 5. Experimental Spectrum for Multiple Layer Geometry.
$H_k - 4\pi M = -1687$ (Left)
$H_k - 4\pi M = -1512$ (Right)

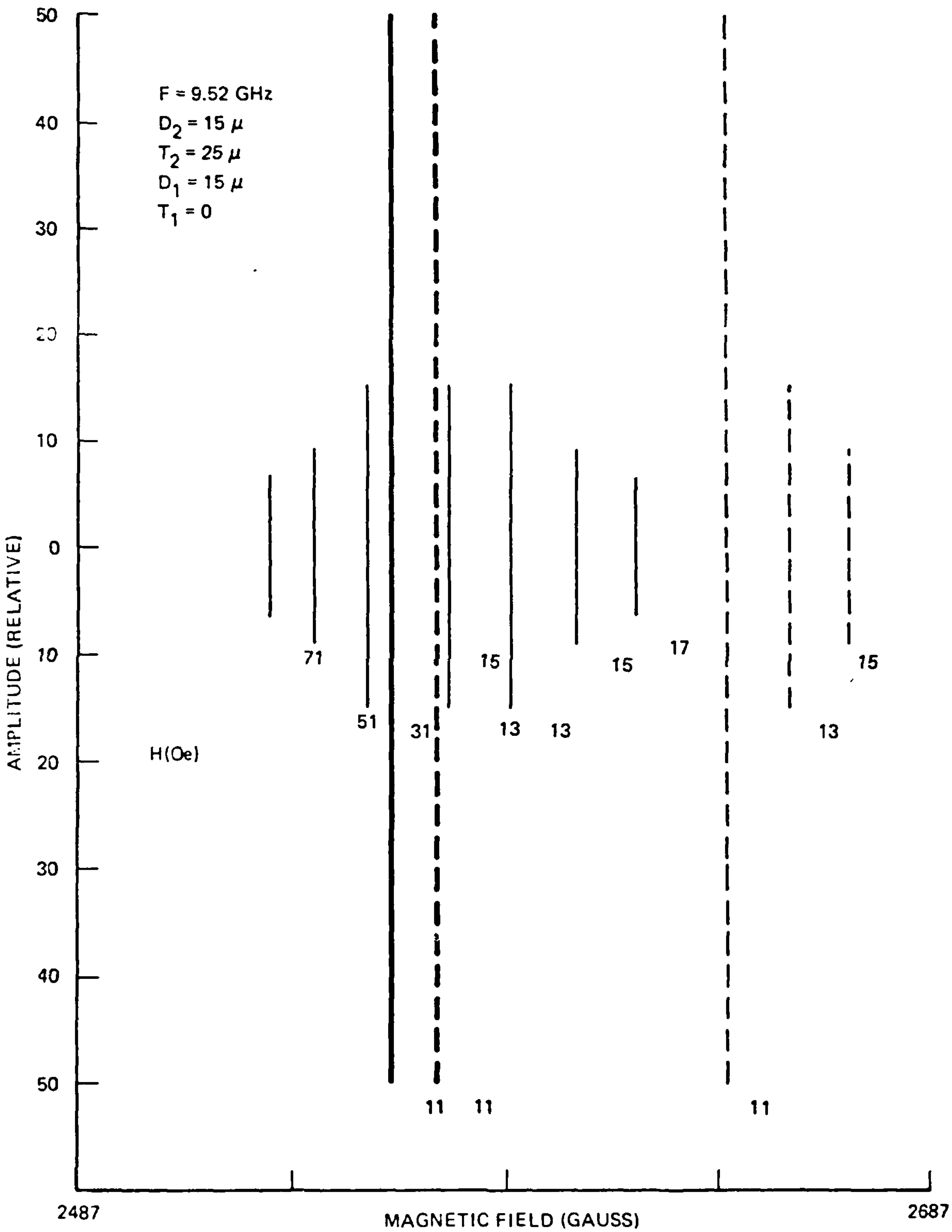

Figure 4. Calculated Spectrum for Multiple Layer Geometry. $4\pi M_1$ = 1512G (Light Broken Lines); $4\pi M_2$ = 1687G (Solid Lines); Coupled Mode (Bold Broken Lines)

Since most FMR spectrometers operate with a fixed frequency and adjustable magnetic field, equation (1) is usually solved with ω given. Then the value of H, which satisfies the equation for any desired combination of m and n, can be calculated. From equation (1), it will be seen that a solution exists corresponding to two isolated single layer films and a solution corresponding to a coupled mode case. Thus, one would expect the spectrum of a sample with this geometry to exhibit a complicated superimposed structure. Further, from energy considerations, it can be expected the m = 0, n = 0 mode will have the largest amplitude and that the amplitude will decrease for the higher order modes (i.e., for larger values of m and n).

A theoretically predicted spectrum and an experimental one are shown in Figures (4) and (5) respectively. Experimentally, the sample consisted of two layers of substitued YIG ($4\pi M_1 \neq 4\pi M_2$) separated by a non-magnetic layer. A similarity between the predicted and experimental spectra is evident, although the two differ considerably in detail. In particular, the coupled mode spectrum is not clear experimentally. This mode is especially important, since it provides the dispersion control required for delay line work. At this state of the investigation, it is possible to interpret the discrepancy between theory and experiment as arising from either an inadequate model or from a less than ideal film.

At the start of the most recent contract year, the main activity on the lithium ferrite task was to continue evaluation of the (111)-oriented epitaxial layers that had been grown during the previous contract year. Those layers had been grown on magnesium indium-gallate spinel substrates from an LPE melt which contained 15.2 mole percent solute (Fe_2O_3 + Li_2CO_3) in a lead oxide-boron oxide solvent. The $PbO:B_2O_3$ mole ratio was 28.6:1.0 and the $Fe_2O_3:Li_2CO_3$ mole ratio was 10.0:1.0. (At the temperature used, 871°C to 884°C, the lithium carbonate decomposes by evolution of CO_2. However, since molar units are used, it does not matter whether we refer to Li_2CO_3 or Li_2O.) Using this melt composition, there were no difficulties with spontaneous nucleation.

sharp interfaces. These points can be illustrated by a brief consideration of the model and its predictions. The current model is a generalization of our multiple layer dispersion relation developed under previous contracts to include the case of finite dimensions (as opposed to semi-infinite). For the case of magnetostatic surface waves (MSSWs) confined to geometry consisting of two magnetic layers separated by a non-magnetic layer, the complete dispersion relation is:

$$\{\bar{\mu}_1\} \quad \{\bar{\mu}_2\} \quad \frac{M_1 M_2}{N^2} + \text{Coth}(-Nt_2)\,[\{\bar{\mu}_1\} - \{\bar{\mu}_2\} - 1] = 0 \quad (1)$$

where

$$\{\mu_\alpha\} = \frac{\mu_\alpha\left[e^{2M_\alpha d_\alpha} - \dfrac{(\mu_\alpha - X_\alpha S_\alpha - N/M_\alpha)}{(\mu_\alpha + X_\alpha S_\alpha + N/M_\alpha)}\right]}{\left[e^{2M_\alpha d_\alpha} + \dfrac{(\mu_\alpha - X_\alpha S_\alpha - N/M_\alpha)}{(\mu_\alpha + X_\alpha S_\alpha + N/M_\alpha)}\right]} - X_\alpha S_\alpha$$

$\alpha = 1, 2$

$\mu_\alpha = 1 - \Omega(\alpha)_H/(\Omega^2(\alpha) - \Omega^2(\alpha)_H)$

$X_\alpha = \Omega(\alpha)/(\Omega^2(\alpha) - \Omega^2(\alpha)_H)$

$\Omega(\alpha)_H = H_o/4\pi M_\alpha(\alpha) = \omega/\gamma 4\pi M_\alpha$

$$M_\alpha = \left[\left(\frac{m\pi}{L}\right)^2 + \left(\frac{n\pi}{W}\right)^2/\mu_\alpha\right]^{1/2}$$

$$N = \left[\left(\frac{m\pi}{L}\right)^2 + \left(\frac{n\pi}{W}\right)^2\right]^{1/2}$$

$m, n = 1, 2, 3, 4 \cdots\cdots$

When ripple suppression techniques were applied to multiple layer LPE YIG delay lines, anomalous characteristics were observed. The passband, instead of exhibiting a smooth variation of loss with frequency, contained several notches. Previously, these had been lost in the ripple. Our theoretical model of magnetostatic wave propagation in multiple layer media does a good job of describing the general characteristics of the passband and delay curves, but does not explain the notches. In order to gain a better understanding of these detailed features, we have been conducting theoretical and experimental studies of ferromagnetic resonance (FMR) in multiple garnet layers. It is hoped that this study will lead to a more accurate description of the real structure of multiple layer LPE garnets. In particular, it should show whether the assumption of sharp interfaces between layers is valid. Also, the FMR analysis should serve as a cross-check on the MSW model since the essential difference between the two cases is that MSW samples are semi-infinite while FMR samples are finite in all dimensions.

If it should be found that the assumption of sharp interfaces is suspect, then several methods of attacking this difficult are available. Earlier in this series of contracts we developed an X-ray diffraction technique for determining the strain and disorder distributions in *nonuniform* thin layers. This work was extended by one of our co-workers to include FMR analysis of such layers and to obtain distributions of the magnetic parameters as a function of depth in the layer. These techniques can be used to characterize the transition regions between layers in multiple layer garnet materials. In addition, we have some control over the structure of transition regions by means of the LPE growth parameters.

At the present time, a model which assumes sharp interfaces gives very good agreement with experiment for the location of those FMR modes which can be attributed to individual layers within multiple layer samples. However, there is not good agreement with experiment for those FMR modes which, according to the model, are composite modes resulting from coupled resonances between layers. Further studies will be performed as part of a new contract (F19628-83-C-0132) to determine whether the discrepancies arise from inadequacy of the model or of the assumptions of uniforms layers with

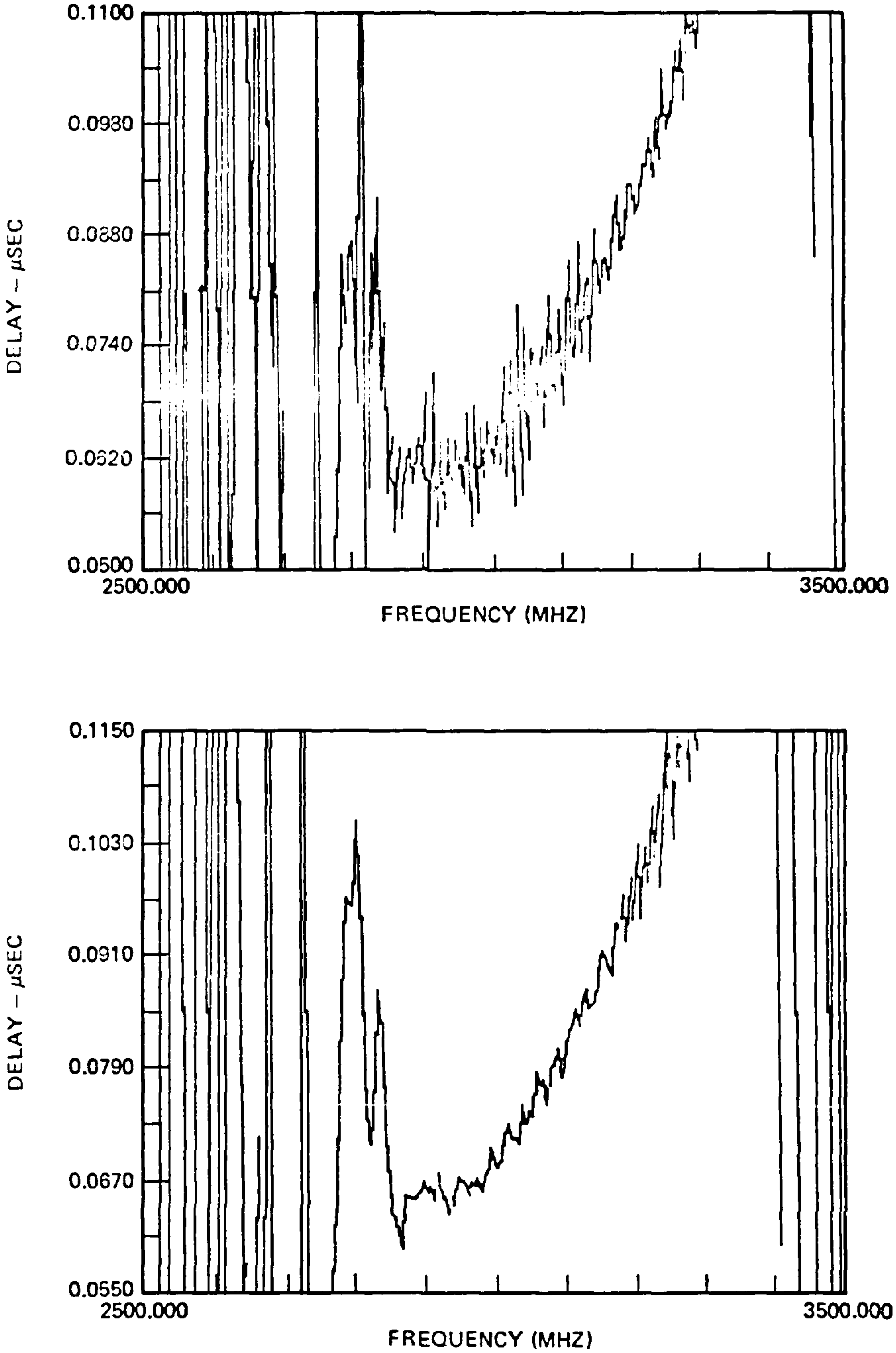

Figure 3. Delay versus Frequency for Magnetostatic Surface Wave Propagation with (Lower Curve) and without (Upper Curve) Ripple Attenuation Layer.

Figure (2) shows the main resonance line of a holmium substituted YIG film. Although the experimental conditions for this measurement were identical to those used to obtain the data in Figure (1), the linewidth is now approximately 140 Oe. The resonance occurs over a field range near that of the original narrow line width material (i.e., the resonance linewidth of the substituted material occurs over a range from 4,948 Oe to 5093 Oe while the unsubstituted YIG has its main resonance at 4879 Oe). Thus, the two major requirements for the attenuation layer, broad line width and similar magnetic properties, are fulfilled by holmium substituted LPE YIG.

We demonstrated this method of ripple suppression using two different configurations. In the first, an ordinary LPE YIG delay line was prepared. This had the form of a rectangular bar comprising a single LPE YIG layer on a GGG substrate. The bar was a few mm wide and about 2 cm long. A network analyzer was used to measure the delay line characteristics. Then the two ends of the bar were dipped, successively, into the holmium-substituted LPE YIG melt to grow layers a few μm thick. Delay line measurements showed about an order of magnitude reduction in ripple.

The second configuration involved growing a uniform layer of holmium-substituted LPE YIG directly on a GGG substrate. By a simple masking and etching step, this attenuation layer was removed from the central area of the wafer. Then an LPE YIG propagation layer was grown on the wafer. The resulting configuration had YIG directly on GGG over the central area and YIG on the attenuation layer near the edge of the wafer. Again, delay line measurements showed substantial suppression of ripple. This is illustrated by the delay vs. frequency curves for magnetostatic delay lines with and without the ripple attenuation layer shown in Figure (3).

These results on ripple suppression, along with further work being pursued under a new contract with RADC Electronic Systems Division (F19628-83-C-0132), serve as a basis for a paper being submitted to the 1984 INTERMAG Conference.

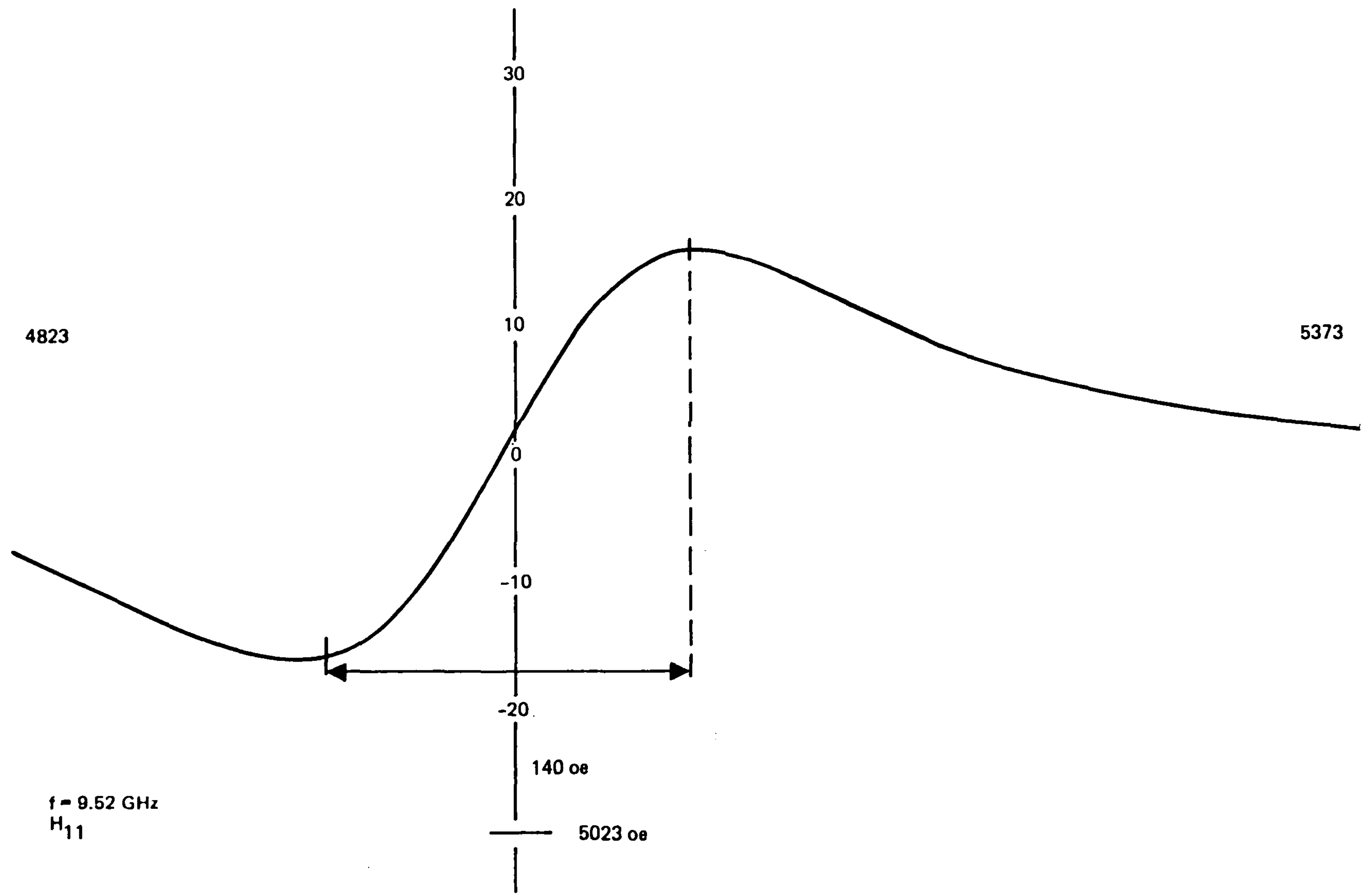

Figure 2. Spectrum of Holmium Substituted YIG on GGG

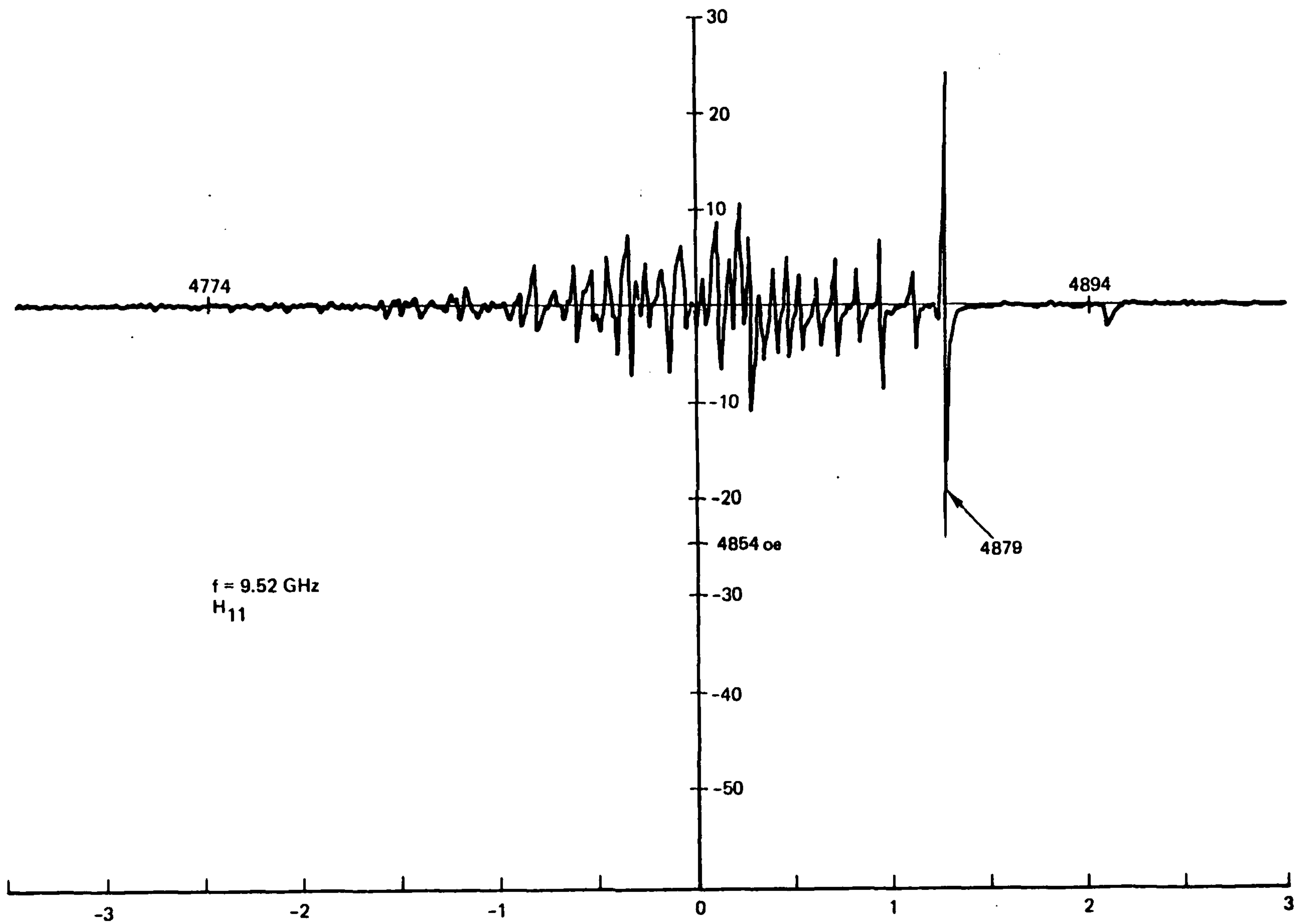

Figure 1. Spectrum of Unsubstituted YIG Film on GGG

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VI SCIENTIFIC INTERACTIONS

Throughout the series of contracts, we have been active participants within the relevent scientific communities: those concerned with magnetism and magnetic materials; crystal growth and characterization; microwave and millimeter-wave ferrite materials and devices. Participation has included conferences and workshops; visits to government, university and industrial laboratories as well as welcoming visitors to our laboratory; supplying samples of our ferrites materials and substrates to researchers at universities and government laboratories. It is gratifying to know that several graduate students have used our materials in their doctoral research. The following paragraphs summarize the scientific interactions during the most recent year of the contract.

The Principal Investigators attended the 3rd Joint INTERMAG-Magnetism and Magnetic Materials Conference in Montreal in July, 1982 where they presented a paper "Forward and Backward Magnetostatic Volume Modes in Multiple Layer Ferrite Films." They also participated in the Magnetostatic Wave Device Technology Workshop at the conference.

Close contacts including laboratory visits were maintained with RADC Electromagnetic Sciences Division at Hanscom AFB and the Army Electronic Devices and Technology Laboratory at Fort Monmouth. Samples of ferrite materials prepared under this contract were supplied to the University of Texas and North Carolina State University, for their research on MSW, and University of California at Irvine for research on optical interactions with magnetostatic waves. L. R. Adkins presented a seminar on MSW devices at Irvine. Samples of the new strontium ferrite material were supplied to Fort Monmouth for evaluation.

H. L. Glass attended the International Conference on Magnetism in Kyoto, partly under contract support, and visited several Japanese industrial and university laboratories engaged in research on epitaxial ferrite materials and MSW devices.

VII INVENTIONS

The following United States Pattents have been issued as a result of inventions made in the course of research on this series of contracts:

1. 4,189,521 "Epitaxial Growth of M-Type Hexagonal Ferrite Films on Spinel Substrates and Composite."

2. 4,200,484 "Method of Fabricating Multiple Layer Composite."

3. 4,243,697 "Self Biased Ferrite Resonators."

4. 4,263,374 "Temperature-Stabilized Low-Loss Ferrite Films."

5. 4,269,651 "Process for Preparing Temperature-Stabilized Low-Loss Ferrite Films."

6. 4,273,610 "Method for Controlling the Resonance Frequency of Yttrium Iron Garnet Films."

7. 4,292,119 "Growth of Single-Crystal $2PbO \cdot Fe_2O_3$."

8. 4,293,372 "Growth of Single-Crystal Magnetoplumbite."

In addition, two applications for United States Patents have been filed as a result of work performed during the past year. The titles are:

9. "Ceramic Strontium Ferrite Borate."

10. "Reduction of Ripple in Magnetostatic Wave Devices."

The last mentioned was reduced to practice under this contract but was conceived under Contract F19628-82-C-0098.

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