Exchange-dominated surface spin waves in thin yttrium-iron-garnet films*

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The study of exchange-dominated nonpropagating surface spin-wave modes observed in thin single-crystal yttrium-iron-garnet films is reported. The angle and temperature dependences of these modes and creation of the surface pinning condition necessary for their existence by the controlled annealing of the films are discussed. The data are consistent with a modified Puszkarski surface-inhomogeneity model in which the surface-anisotropy field has a tensorial form. At the substrate surface the in-plane component of this field, $K_{\parallel}^{s}$, is large and negative and the perpendicular component, $K_{\perp}^{s}$, is large and positive. The degree of localization varies with the annealing history of the sample, and etching experiments confirm a decay length as small as 500 Å when the magnetization lies in the plane of the film. As the magnetization is rotated out of the film plane, a "critical angle" at which the uniform mode is excited is observed, and at the perpendicular orientation the surface mode has been transformed to the first body mode of the film. With decreasing temperature, $K_{\parallel}^{s}$ is observed to decrease in magnitude and pass through zero between 100 and 200 K. At 4 K, $K_{\parallel}^{s}$ is large and positive. These data suggest that a psuedodipolar exchange interaction may be occurring between the Fe and Gd atoms at the substrate interface. By annealing at 1200 °C, a similar surface effect is produced at the free surface of the film having $K_{\parallel}^{f} < 0$ and $K_{\perp}^{f} > 0$. However, the temperature dependence of the free-surface anisotropy field indicates that the origin of the interaction has a very different microscopic basis.

I. INTRODUCTION

The excitation of nonpropagating surface spin-wave modes has recently been observed in single-crystal yttrium-iron-garnet (YIG) films.[1-4] These exchange-dominated surface modes are different from magnetostatic surface modes,[5-7] which generally are long-wavelength modes having negligible exchange energies. Also, the magnetostatic surface modes have a real wave vector in the plane of the film and an imaginary component normal to the plane, with a characteristic decay length often many times the film thickness. As a result, the magnetostatic surface modes are propagating waves and in finite samples their normal-mode resonant frequencies depend on the geometry of the film. The exchange surface modes reported here have an imaginary wave vector $k_z = i\delta$ normal to the surface of the film and a zero component in the plane of the film. Values of $\delta^{-1} = 500$ Å have been observed. These exchange surface modes are nonpropagating and independent of the shape of the film.

There are two possible branches of exchange surface waves in the spin-wave spectrum. The surface wave having a pure imaginary propagation vector $k_z = i\delta$ is called an acoustic surface spin wave, while the surface wave having a complex propagation constant $k_z = \pi/a + i\delta$, where a is the lattice constant, is called an optical surface spin wave.[8] Thus an acoustic spin wave lies below the bulk spin-wave energy band while the optical spin wave lies above the spin-wave energy band. In a spin-wave-resonance (SWR) spectrum, the resonance-field position of an acoustical spin-wave mode is above that of the uniform precession mode and the field position of an optical surface spin-wave mode is the lowest in samples of finite thickness. The reports of experimental observations of exchange surface spin-wave modes have been restricted to the acoustic type.

The spin-wave resonance modes excited in thin magnetic films depend on the surface boundary conditions. The origin of these surface boundary conditions has been a subject of both theoretical and experimental investigations for some time. In a series of papers, Sparks[9] has considered a variety of internal field conditions and surface parameters to give the positions, intensities, and linewidths of the magnetoexchange branches in insulator films. Most of the analyses have been carried out for "perpendicular resonance" (the magnetic field oriented perpendicular to the film plane[10-12]) or parallel resonance.[13] Very little has been reported at intermediate angles.

Only recently has the role of the nonpropagating surface mode as a normal mode in the spin-wave spectrum received special emphasis.[8,14,15] Puszkarski,[8] in particular, proposes a surface anisotropy field present at the film surfaces that allows the excitation of a nonpropagating surface mode and predicts certain angular dependences of the surface-mode characteristics. With a few modifications, this surface-inhomogeneity model has been quite successful in predicting the many unique properties of nonpropagating surface modes.

In addition to the perpendicular orientation, this

report includes SWR experiments that have been performed at parallel resonance and intermediate angle configurations. The SWR spectrum in YIG films is observed to depend on the magnetic field orientation in a manner similar to that reported previously in metallic films.[16] However, the excellent agreement between the magnetoexchange dispersion relations observed experimentally and derived theoretically indicate that the internal fields in the YIG films are very homogeneous and that the origin of the surface exchange mode reported here is due to a surface effect and not a volume inhomogeneity, as observed in the metallic films. A unique feature reported here is that the spin waves excited in these YIG films are observed to be tunable in that the wavelength of the spin-wave modes is observed to change as the direction of the applied magnetic field is rotated with respect to the direction normal to the film. In other words, the propagation constant of the spin-wave modes excited in YIG films is orientation dependent, and the evolution of the nature of the modes, particularly that of the exchange surface mode, is found to agree with a modification of Puszkarski's model.[2,3,8]

In order to understand the conditions necessary for the existence of the surface modes, a number of YIG films have been annealed in an oxygen atmosphere to study the correlation between the annealing conditions and the occurrence of surface modes. Experimental results indicate that the surface conditions are strongly influenced by the annealing process.

To provide a theoretical background for this paper, Puszkarski's surface-inhomogeneity model of the spin-wave resonance modes and the spin-wave dispersion relation with cubic magnetocrystalline anisotropy included are reviewed in Sec. II. Section III is a detailed report of the experimental results which confirm the excitation of the nonpropagating surface exchange mode. The angle dependence of the spin-wave modes, the effects of the annealing of the films on the spin-wave spectra, the results of chemical etching to determine the degree of localization of the surface modes, and the temperature dependence of the surface modes are also reported.

In Sec. IV, a detailed comparison of the experimental results is made with the modified Puszkarski surface-inhomogeneity model. Possible origins of the surface anisotropy are discussed.

II. THEORY

A. Puszkarski's surface-inhomogeneity model

The normal spin-wave modes excited in a thin magnetic film, in Puszkarski's calculations,[8,15] are the solutions of a Hamiltonian that includes the nearest-neighbor Heisenberg exchange interaction, a generalized Zeeman interaction including the effects of the applied magnetic field, the shape demagnetization field, and an effective surface anisotropy field. The latter, acting on the surface spins, is assumed to exist at each surface of the sample. The pinning condition at each film surface can be described by a "surface parameter" A given by

$$A = 1 - (g\mu_B/2SzJ)\,(\vec{K}_s \cdot \hat{\gamma}) \ , \tag{1}$$

where g is the spectroscopic-splitting factor, μ_B is the Bohr magneton, S is the spin of the atoms considered, z is the number of nearest neighbors, J is the Heisenberg interaction constant, $\hat{\gamma}$ is a unit vector parallel to the direction of the magnetization, and $\vec{K}_s$ is the surface anisotropy field acting on the surface spins. In essence the surface parameter is 1 minus the ratio of the surface anisotropy field to the bulk Heisenberg exchange field. The surface spin is defined as "pinned" if $A < 1$ and "natural" if $A = 1$. These definitions can be compared with the conventional pinning conditions[9] at a film surface

$$am^+ + b\,\frac{\partial m^+}{\partial z} = 0 \ . \tag{2}$$

The condition $b = 0$ is equivalent to the condition $A = -\infty$, while the condition $a = 0$ is equivalent to the condition $A = 1$. However, there is no equivalent condition implied in Eq. (2) for the condition $A > 1$. However, this is the condition for which the surface modes are permitted.

For symmetrical boundary conditions, where the surface parameters at both surfaces are identical, a single parameter is sufficient to describe the normal spin-wave modes. When $A < 1$, only body spin-wave modes having real values of the wave vectors can be excited. When $A > 1$, the surface spin has lower energy than the bulk spins, and the acoustic-type surface modes as well as the body modes can be excited. When $A = 1$, all spins throughout the sample have the same effective field, and the uniform-precession mode $k = 0$ is the only normal mode that will be excited by a uniform microwave field in the SWR experiment.

As a function of the surface parameter A, the first ($n = 1$) and highest field mode in a SWR spectrum is an acoustic-type surface mode when $A > 1$. The degree of localization of the surface mode decreases as A decreases in magnitude. If A is decreased to unity, the surface mode becomes the uniform-precession mode. The uniform-precession mode transforms into the first of the body modes as A further decreases in magnitude and approaches $k_1 = \pi/L$, or a half-wavelength mode as A approaches $-\infty$. This evolution of the normal spin-wave modes

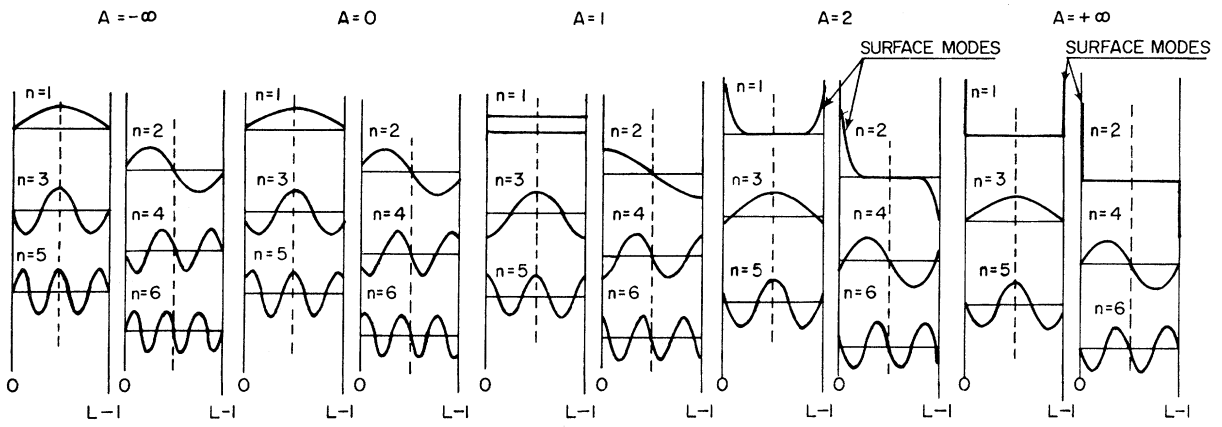

FIG. 1. Shapes of spin-wave modes (with low n) for various values of the surface parameter A. Symmetric and antisymmetric modes correspond to odd and even n, respectively.

as a function of the surface parameter is shown in Fig. 1.[8,17] The relative intensities of the first few normal modes for various values of the surface parameter is shown in Fig. 2.[8] When $A=-\infty$ the surface spins are completely "pinned" and the normal modes are

$$k_n = n\pi/L, \quad n = 1, 2, 3, \ldots , \tag{3}$$

where k_n is the wave vector of the nth mode and L is the film thickness. For this case the separation between the resonance field of any two body modes, say the pth and the rth modes, is equal to $(r^2-p^2) \times D(\pi/L)^2$, and the quadratic law is obeyed. This quadratic relation is often used as the basis for the experimental determination of the exchange constant D from the SWR data. However, when $-\infty < A < 1$, the wave vectors of the modes deviate from the relation in Eq. (3). This deviation is taken into account in Puszkarski's model, where the wave vector is expressed as $k_n = (n-\Delta)\pi/L$, $n = 1, 2, 3, \ldots$, where Δ is a measure of the amount of deviation from the normal modes defined in Eq. (3). In general Δ depends on the mode number n, the pinning parameter A, and the film thickness L. For relatively thick films, it is possible that Δ is a linear function of the mode number. Assuming that $\Delta = \lambda n$, where $\lambda < 1$, $n = 1, 2, 3, \ldots$, these modes still fit a quadratic relationship, and the apparent exchange constant so determined would differ from the true exchange constant by a factor of $(1-\lambda)^2$.

Puszkarski has considered the case of a unidirectional surface anisotropy field K_s having an arbitrary magnitude and an arbitrary orientation with respect to the normal direction of the film. The surface parameter is given by

$$A = 1 - (g\mu_B K_s/2SzJ)\cos(\alpha_0 - \phi) , \tag{4}$$

where α_0 is the angle between the orientation of the unidirectional surface anisotropy field and the normal direction, and ϕ is the angle between the unit vector $\hat{\gamma}$ and the normal direction. Therefore the normal spin-wave modes depend on the orientation of the applied magnetic field with respect to the normal of the film. The critical-angle orientations at which $A = 1$, i.e., the orientations for which the magnetization is perpendicular to the unidirectional surface anisotropy field, are given by $\phi_c = \frac{1}{2}\pi + \alpha_0$, according to Eq. (4). For this model

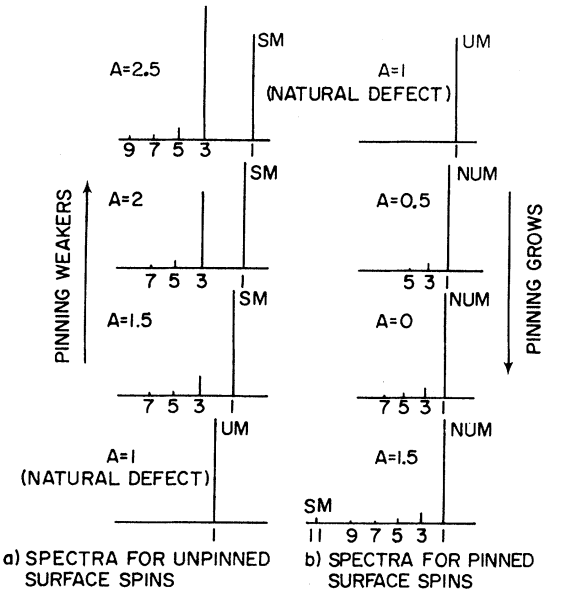

FIG. 2. SWR spectra calculated for various values of the surface parameter A, i.e., for various pinning of the surface spins. The film has 11 spin layers. UM denotes a uniform mode, SM a surface mode, NUM a nonuniform body mode.

the critical angles are expected at only two orientations of the applied field which differ by π rad.

Experimentally, a critical angle, where only a single mode is observed, does exist in our samples. However, four such angle positions are observed in a rotation of 360° instead of the two predicted above. The critical angles are found to have a conical symmetry about the normal direction of the film as indicated in Fig. 3.

It is possible to include this conical symmetry in Puszkarski's model by introducing a tensorial form of the surface anisotropy field, namely,

$$\underline{K} = \begin{bmatrix} K_\perp & 0 & 0 \\ 0 & K_\parallel & 0 \\ 0 & 0 & K_\parallel \end{bmatrix}, \tag{5}$$

where $K_\perp$ is the component of the anisotropy field tensor when the magnetization is normal to the film surface and $K_\parallel$ is the in-plane component of the field tensor. The surface parameter then takes the form

$$\begin{aligned} A &= 1 - (g\mu_B/2SzJ)(\hat{\gamma}\cdot\underline{K}\cdot\hat{\gamma}) \\ &= 1 - (g\mu_B/2SzJ)(K_\perp\cos^2\phi + K_\parallel\sin^2\phi)\,. \end{aligned} \tag{6}$$

The condition for the critical angle ϕ_c where $A = 1$, is given by

$$\tan\phi_c = (-K_\perp/K_\parallel)^{1/2}\,. \tag{7}$$

A critical angle is thus possible if $K_\parallel$ and $K_\perp$ are of opposite sign. For a surface anisotropy having $K_\parallel < 0$ and $K_\perp > 0$, a surface mode (acoustic type) exists in the region $\phi_c < \phi < 90°$, and only body modes exist in the region $0 \leq \phi < \phi_c$, as indicated in Fig. 3.

For asymmetrical surface boundary conditions, two independent surface parameters are necessary to describe the normal spin-wave modes. It is thus more difficult to analyze such a case quantitatively. However, most qualitative results can be carried over from the symmetrical-boundary-condition case to the asymmetrical case. One or two acoustic-type surface exchange modes can be observed in an asymmetrical system depending on the strength of the two surface anisotropy fields. For relatively thick films, i.e., films having a thickness much greater than the nearest-neighbor distance, the conditions[15] for the existence of a single acoustic surface mode in a SWR spectrum is that one surface parameter is greater than unity and the other less than or equal to unity. This surface mode is again the highest field mode in a SWR spectrum.

When both surface parameters are greater than one and unequal, two acoustic surface modes can be excited, with the highest-field mode corresponding to a quasisymmetric surface mode and the next mode corresponding to a quasiantisymmetric surface mode. For the quasisymmetric surface mode the amplitude of the oscillating dipole moment is greatest at the surface having the stronger surface anisotropy field.[15] In such a case the surface mode is said to be localized at this surface. For the quasiantisymmetric surface mode the amplitude is larger at the surface having the weaker surface anisotropy field.[15]

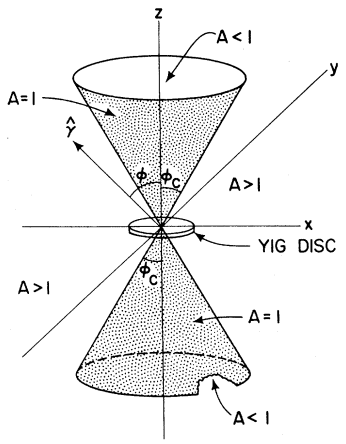

FIG. 3. Angle dependence of the surface parameter A for a tensorial surface anisotropy energy.

B. Spin-wave dispersion relation

The dispersion relation for long-wavelength spin waves in a single-crystal ferromagnetic film, including the applied magnetic field, the shape demagnetization field, the magnetocrystalline field, and the Heisenberg exchange, is given by[18]

$$\begin{aligned} (\omega/\gamma)^2 = &[H_0\cos(\phi-\theta) - 4\pi M\cos^2\phi - g_1(\phi) + Dk^2] \\ &\times[H_0\cos(\phi-\theta) - 4\pi M\cos^2\phi - g_2(\phi) + Dk^2]\,. \end{aligned} \tag{8}$$

The equilibrium orientation of the saturation magnetization is determined by

$$2\pi M\sin 2\phi - H_0\sin(\phi-\theta) + g_3(\phi) = 0\,, \tag{9}$$

where ω is the angular frequency of the spin wave, γ the gyromagnetic ratio, H_0 the strength of the applied magnetic field, $4\pi M$ the demagnetization field, D is the exchange constant, k is the spin-wave wave vector, θ is the angle between the direction of the applied magnetic field and the film normal, ϕ is the angle between the direction of the saturation magnetization and the film normal, and g_1, g_2, and g_3 are magnetocrystalline fields which in general depend on the crystal symmetry, the orientation of the film, and the crystallographic plane in which the applied magnetic field is located. For a [100]-oriented YIG film, with the applied magnetic field in the (100) plane, the magnetocrys-

talline fields have the form

$$g_1(\phi) = -H_K \cos 4\phi \ , \tag{10a}$$

$$g_2(\phi) = -\tfrac{1}{4} H_K (3 + \cos 4\phi) \ , \tag{10b}$$

$$g_3(\phi) = \tfrac{1}{4} H_K \sin 4\phi \ , \tag{10c}$$

where $H_K = 2|K_1|/M$ is the first-order cubic crystalline field. These expressions include the negative sign of K_1 for YIG. The application of Eqs. (8)–(10) to specific resonance data requires that values of H_K and $4\pi M$ be known. Since the precision of these values is less than the magnetostatic contribution and the stress effects in thin YIG films have the same angle dependence as the demagnetization field,[19] the latter have not been included. The uniform precession field ($k = 0$) can be determined from the two simultaneous equations (8) and (9), and the result for a [100]-oriented film with the geometrical configuration specified above for Eq. (10) is given by

$$H_u(\theta) = \frac{2\pi M}{\cos(\phi - \theta)} \left\{ \frac{1 + 3\cos 2\phi}{2} + \frac{H_K}{4\pi M} \left(\frac{3 + 5\cos 4\phi}{4} \right) + \left[\left(\frac{\cos 2\phi - 1}{2} + \frac{H_K}{4\pi M} \frac{3(\cos 4\phi - 1)}{4} \right)^2 + \left(\frac{\omega}{4\pi\gamma M} \right)^2 \right]^{1/2} \right\} , \tag{11}$$

where the magnetization angle ϕ can be evaluated as a function of the magnet angle θ by substituting the expression for H_0 obtained from Eq. (9). The resonance field for a spin-wave mode can also be obtained from Eq. (8), and the result is given by

$$H_n = H_u - \frac{Dk_n^2}{\cos(\phi - \theta)} \ , \tag{12}$$

where the spin wave vector k_n theoretically has to be determined from the characteristic equations,[8] which in turn depend on the surface parameters. (See Fig. 1).

When the applied magnetic field is perpendicular to the film surface, the resonance field positions for the spin-wave modes, obtained from Eqs. (11) and (12), are given by

$$H_n = \omega/\gamma + 4\pi M + H_K - Dk_n^2 \ . \tag{13}$$

Generally speaking, it is very difficult to determine the physical parameters like γ, $4\pi M$, H_K, and D to a desirable degree of accuracy from the SWR analysis alone. The primary reason is the difficulty in determining the exact values of the normal wave vectors k_n from first principles. The common practice is to assume that the observed modes satisfy the perfectly dimensional resonance conditions [Eq. (3)] and determine if the quadratic relation is valid or not for the observed modes. If the quadratic relation holds, the saturation magnetization is deemed to be uniform and the exchange constant can then be determined from the slope of a graph of H_n vs n^2, provided the thickness of the sample is known. A significant deviation from the quadratic law may signify the existence of an inhomogeneity in the internal distribution of either the saturation magnetization, magnetocrystalline field, demagnetization field, or stress.[20–22] If this is the case, it is difficult to determine the exchange constant and other parameters to any great degree of accuracy. Even if the quadratic relation is observed to hold, it is still not completely certain that the perfectly pinned condition is satisfied. In addition to the quadratic dispersion relation, the relative intensity of the modes must also obey the $1/n^2$ law. But it is also difficult to determine very accurately from SWR spectrum the relative intensities of the observed modes. In general, the deviation from the dimensional resonances is different for each individual mode.[8,21,22]

III. EXPERIMENTAL RESULTS

A. Experimental procedure

The single-crystal YIG (Y_3FeO_{12}) films used in these experiments were grown on GdGaG ($Gd_3Ga_5O_{12}$) substrates at the Electronics Research Div. of Rockwell International, using chemical-vapor-deposition (CVD) techniques.[23] These crystals are of high quality both optically and magnetically. The agreement between theory and experiment for the magnetoexchange-branch dispersion relations[11] is a confirmation of the good magnetic properties of the films. The linewidth of the spin-wave modes observed with these films is approximately 1 Oe. The thickness of the YIG films varies from 0.37 to 1.15 μm, and [100]-, [110]-, and [111]-oriented films were used. The microwave spectrometer used in this experiment is a standard reflection type operating at either 9, 23, or 34 GHz frequency range. The SWR spectrum is detected as the first derivative of the absorption signal, using a 400-Hz modulation field. An Andonian model MDH-3L double-walled variable-temperature Dewar was used to control the sample temperature to ± 0.5 K over the range 300–2 K. A Magnion model G-502 NMR precession gaussmeter was used to calibrate the field strength of a 15-in. Varian electromagnet. The strength of the magnetic field can be accurately measured to within 1

Oe, and the magnet angle θ can be measured to within $\frac{1}{2}°$.

B. Evidence confirming a surface mode

The first three high-field modes of a typical YIG-film sample showing the presence of an acoustic-type surface exchange mode in the SWR spectrum are shown in Fig. 4. The applied magnetic field is directed along a [010] direction in the surface of a [100]-oriented film. The spectrum is taken at 9.16 GHz and at room temperature. The sample is disk shaped and has a thickness of 0.56 μm. The theoretical field value of the uniform mode can be estimated[24] and is shown as an arrow (UM) in Fig. 4. The weak high-field mode which is observed 85 Oe above the uniform mode can be identified as an acoustic-type surface exchange mode. The resonance-field position of this mode is independent of the diameter of the films from the same deposition run, or of the shape of the film. This indicates that no significant magnetostatic energy contributes to this surface mode. The existence of an acoustic-type surface exchange mode can be identified from the SWR spectrum by the following experimental facts: (i) the resonance field of the mode is above the theoretical uniform-mode field position; (ii) the intensity of the mode may be equal to or smaller than the next-lower field mode; (iii) the angle dependence of this mode is consistent with the predictions for a surface mode as given by Puszkarski (to be discussed in Sec. IIC); and (iv) the intensity of the mode as a function of the film thickness is consistent with that of a surface mode (to be discussed in IIE). Metallization of the outside surface of the film has little effect on the intensity of the surface mode, indicating that this surface mode is a nonpropagating mode localized at the substrate surface.[1]

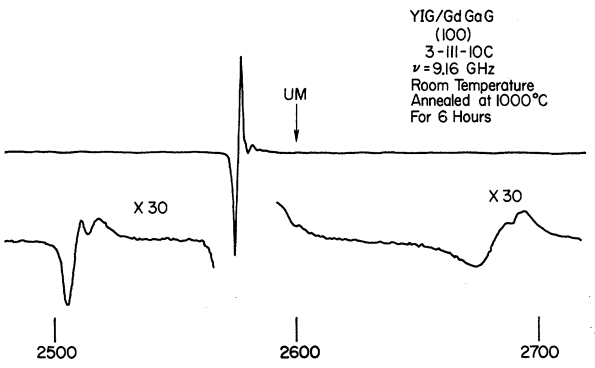

FIG. 4. Trace of the high-field modes at parallel resonance, 9.2 GHz and room temperature for a YIG film annealed at 1000 °C in an oxygen atmosphere for 6 h.

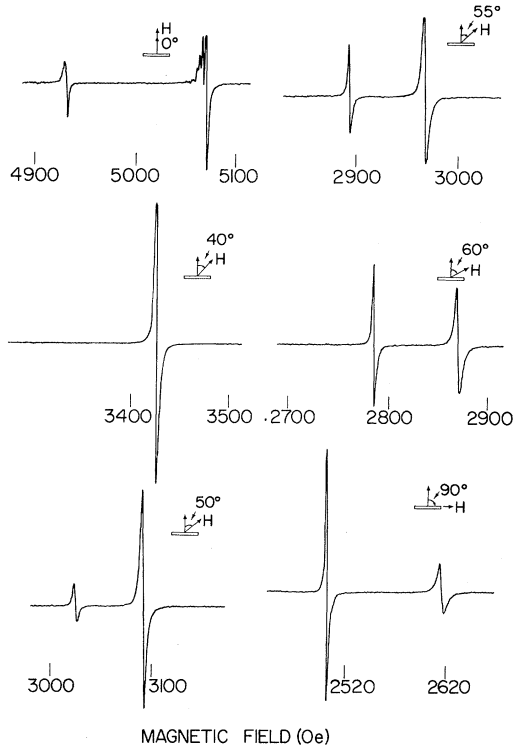

FIG. 5. Trace of the first two modes observed at six orientations at 9.2 GHz and room temperature for a typical YIG film having a single surface mode.

C. Angle dependence of the surface mode

Since the surface anisotropy energy depends generally on the orientation of the saturation magnetization, the surface parameter A is a function of the magnetization angle ϕ as represented by Eq. (6). As a result, the nature of the normal spin-wave modes excited in a film is angle dependent. This angle-dependence phenomenon which is due to the anisotropic nature of the surface pinning conditions has been observed in all YIG samples used in these experiments.

For example, the angle dependence of the mode positions and relative intensities are shown for a series of spectra reproduced in Fig. 5. The data were taken for a 0.37 μm thick film at 9.16 GHz and at room temperature. Only the first two high field modes observed are shown. The weak high-field mode observed at parallel resonance is identified as a surface mode while the next and strongest mode is the first of the body modes. As the applied magnetic field is rotated towards the perpendicular orientation, the high-field surface mode is observed to increase in intensity while the body modes all decreases in intensity. At the critical angle, which is $\theta_c = 40°$ for this film, the surface mode becomes the only mode observed and corre-

sponds to the uniform precession mode, while the higher-order modes are not observed.

Beyond this critical angle, the surface mode transforms into the first of the body modes and the higher-order body modes are again observed as the magnetic field is rotated towards the perpendicular direction. The angle dependence of the SWR spectra shown in Fig. 5 is consistent with the modified Puszkarski model as represented in Fig. 2. In summary, when a single surface mode is observed in these YIG films, the surface mode is observed in the region $\theta_c < \theta \leq 90°$; the surface mode becomes the uniform mode at $\theta = \theta_c$; and the uniform mode transforms into the first body mode in the region $0° \leq \theta < \theta_c$.

The resonant-field separation between the two highest-field modes is observed to change with the magnet angle θ, as shown in Fig. 6. The minimum separation occurs near the critical angle ($\theta_c = 40°$ in Fig. 6). At the critical angle the higher-order modes become symmetric across the film and are not excited by a uniform microwave field.

These results also substantiate the assumption of a nearly complete pinning condition at perpendicular resonance and an unpinned condition at the critical angle. Assuming that the highest-field mode makes a transition from an $n = \frac{1}{2}$ mode at perpendicular resonance to the uniform mode at θ_c and that the second mode goes from an $n = \frac{3}{2}$ mode to an $n = 1$ mode, then Eq. (12) can be used to show that the ratio of the mode separation at the perpendicular orientation and the critical angle will be 2.

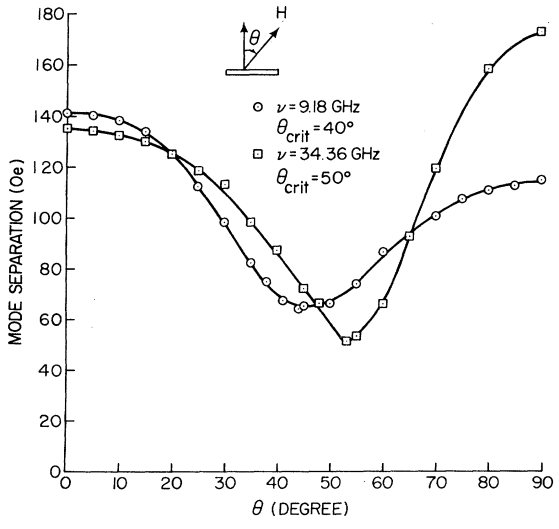

FIG. 6. Angle dependence of the resonant-field separation between the two highest field modes at 9.2 and 34.4 GHz and room temperature for a typical YIG film having a single surface mode.

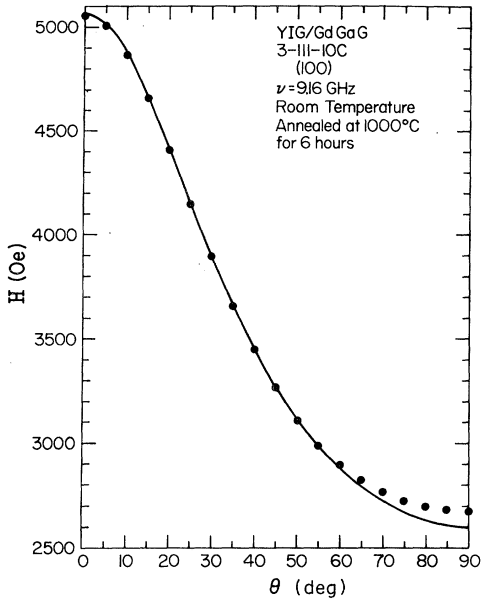

FIG. 7. Angle dependence of the resonant-field position of the highest field mode of a typical YIG film having a single surface mode.

The ratio of the experimentally observed separations of approximately 2 is thus good evidence of nearly complete pinning at the perpendicular orientation.

Figure 7 shows the positions of the resonance field of the high-field mode for a YIG film showing the presence of a surface mode. The data is taken for a 0.56-μm [100]-oriented film at 9.16 GHz and at room temperature. At parallel resonance the applied magnetic field is arranged to be along the direction of a [010] cubic axis, so that the static field is rotated in a (001) plane as it moves from the parallel orientation to the perpendicular orientation. These orientations of the static field fit the conditions for the application of Eq. (11). The solid line in Fig. 7 is the calculated uniform field position of the sample using Eq. (11). The values $(4\pi M)_{\text{eff}} = 1722$ Oe and $H_K = 82$ Oe have been used in this calculation, with the former defined by $(4\pi M)_{\text{eff}} = 4\pi M_s - H_\sigma$, where H_σ is the magnetostrictive anisotropy field.[19]

The high-field mode which is a surface mode near the parallel orientation is observed to resonate above the calculated positions of the uniform mode. Near the perpendicular direction, the high-field mode, which has transformed into a body mode in this region, is observed to resonate below the calculated position of the uniform mode. At the critical angle the observed resonant field for the uniform mode is found to fall within 15 Oe of the calculated value for all samples. This indicates that

a good degree of accuracy has been obtained in the estimation of $4\pi M$ and the alignment of the orientation of the static field with respect to the sample.

The angle dependence of the magnetic field separation between the calculated value of the uniform mode and the spin-wave modes is shown for a typical film in Fig. 8. The experimental positions of the first three high-field modes relative to the calculated values of the uniform mode is plotted as a function of the magnetic field angle θ. The film thickness is 0.56 μm. The field difference $H_u - H_n$, according to Eq. (12), is $Dk_n^2/\cos(\phi-\theta)$. At the microwave frequency ($\nu = 9.16$ GHz) employed and the values of the saturation magnetization of YIG, $4\pi M \sim 1750$ Oe, the difference between the magnetization angle and the magnetic field angle is found to be $0° < \theta - \phi < 15°$; therefore $\cos(\phi-\theta) \simeq 1$. Since the Fe^{3+} magnetic ions are S-state ions in $Y_3Fe_5O_{12}$ the exchange constant D is not expected to have any significant anisotropy. Figure 8 is therefore a plot of the experimental values of k_n^2 of the normal modes as a function of the angle θ. Negative values of k_1^2 indicate that the wave vector k_1 is pure imaginary, characteristic of a surface mode. The solid lines are the calculated positions of the normal modes $k_n = n\pi/L$, $n = 1, 2, 3, \ldots$, for $L = 0.56$ μm, which corresponds to the case when the surface spins are completely pinned at each surface, i.e., $A = -\infty$. It is included for the sole purpose of indicating the positions of the observed modes relative to the dimensional-resonance modes. The bulk value of the exchange constant of YIG,[25] $D = 0.516\times10^{-8}$ Oe cm^2, was used in the calculation. Experimental evidence like that of Fig. 8 confirms that the normal spin-wave modes excited in a magnetic film are in general dependent on the orientation of the magnetization with respect to the film normal.

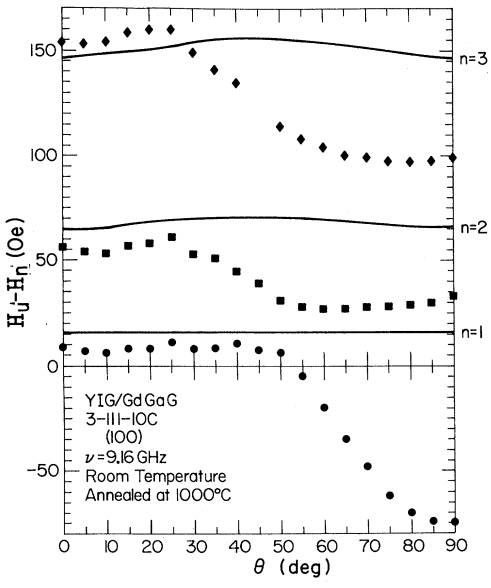

FIG. 8. Angle dependence of the magnetic field separation of the observed positions of the spin-wave modes H_n from the calculated position of the uniform mode H_u for a typical YIG film having a single surface mode.

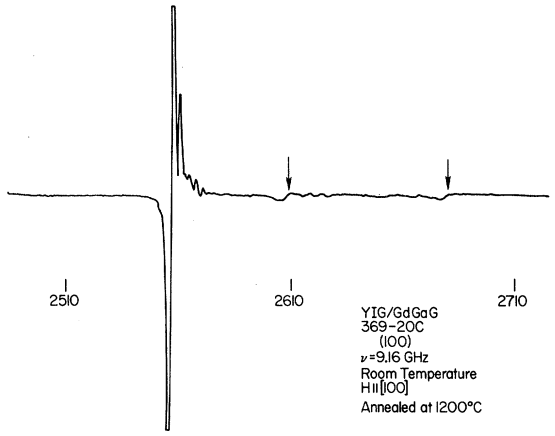

FIG. 9. Trace of the high-field modes observed at parallel resonance, 9.2 GHz, and room temperature for a YIG film annealed at 1200 °C in an oxygen atmosphere for 6 h.

Two surface exchange modes which are observed to resonate above the uniform-field position have also been observed in several YIG films. Figure 9 shows a typical case when two surface modes are observed at parallel resonance. The highest field mode is a quasisymmetric surface mode and the second high-field mode is a quasiantisymmetric surface mode. The angle dependence of the two surface modes is shown for a 0.56-μm-thick sample in Fig. 10. At the parallel orientation both surface modes are observed above the uniform-field position. The quasisymmetric surface mode transforms into the first body mode, which is nearly a half-wavelength mode, at the perpendicular orientation. The first body mode, which is the strongest mode observed at parallel orientation, transforms into the third body mode, which is nearly a three-halves wavelength mode, at the perpendicular orientation. The solid lines correspond to the $n = 0$, π/L, $2\pi/L$, and $3\pi/L$ modes, using the bulk value of the exchange constant.

D. Effects of annealing

Since surface modes have not been observed in all YIG samples studied, a detailed investigation of the SWR spectrum of a number of samples systematically annealed in this laboratory has been undertaken. As-grown samples have been annealed in dry oxygen atmosphere at temperatures between

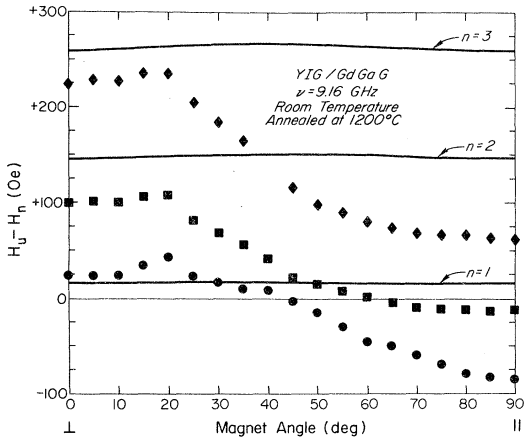

FIG. 10. Angle dependence of the magnetic field separation of the observed positions of the spin-wave modes H_n from the calculated position of the uniform mode H_u for a typical YIG film having two surface modes.

600 and 1200 °C for 6 h. Primarily, two series of YIG films having thicknesses of 0.56 and 1.15 μm, respectively, have been used in this study. The samples of each series are from the same deposition run, and the SWR spectra are nearly identical before annealing. The SWR spectra obtained from the annealed samples indicate that the annealing temperature is a more important factor in effecting a change in the spectra than the duration of annealing. Furthermore, the results show that the properties of the surface exchange modes are closely related to the annealing temperature. For films annealed above 725 °C, a single surface mode can be readily identified in the parallel resonance spectrum.

The stick diagrams in Fig. 11 represent the parallel resonance spectra for a series of samples 1.15 μm thick which are either unannealed or annealed at 740, 900, 1000, 1100, and 1200 °C. Each line represents an observed signal, and the height is an indication of the relative intensity as compared to the other signals observed from the same film. The horizontal scale represents the absolute values of the resonant magnetic fields. The highest field mode observed in the samples annealed at 740, 900, 1000, and 1100 °C can be easily identified as a surface exchange mode. This mode is observed above the calculated uniform-field position and its intensity is less than that of the next-lower field mode which is the strongest body mode. This surface exchange mode becomes the only spin-wave mode observed at the critical angle and transforms into the first and strongest body mode at perpendicular resonance.

The highest field mode of the spectrum of the unannealed film is also a surface mode, as evidenced by the single mode observed at the critical angle. However its location near the value of H_u and the high intensity of this mode indicates that it is only slightly localized. As the annealing temperature is increased from 740 to 1100 °C, the single surface mode observed in each film is detected at a higher resonance-field position, indicating a higher degree of localization at the substrate interface.

For the film annealed at 1200 °C, two weak high-field signals can be identified as surface modes, and the angle dependence of those modes is shown in Fig. 10. For as-grown samples, both odd-ordered and even-ordered modes are observed in the SWR spectra. At perpendicular resonance these modes are nearly equal to the $k = n\pi/L$, $n = 1, 2, 3, \ldots$, modes. The most significant change with regard to the body modes observed at perpendicular resonance is that the even-ordered modes decrease in intensity with respect to the odd-ordered modes as the annealing temperature is increased above 1000 °C. The even-ordered modes are not observed for some of the films annealed at 1200 °C.

As to the bulk properties of the film, the dominant first-order cubic crystalline anisotropy field having an empirical value of −82 Oe remains unchanged upon annealing. The value of the saturation magnetization which is estimated by assuming the quadratic fit of the perpendicular resonance modes also remains relatively unchanged upon annealing.

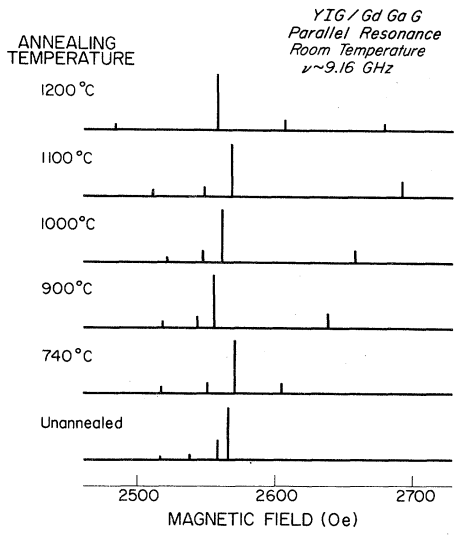

FIG. 11. Stick diagrams representing the parallel resonance spectra of a series of YIG films showing the effect of annealing on the resonant-field position of the spin-wave modes.

E. Localization of surface modes

As a function of the film thickness, the signal intensity and resonance-field value of surface modes and body modes are different. For body modes, the distribution of the amplitude of the oscillating dipole moment is trigonometric across the thickness of the film, and the resonance field and signal intensity is expected to decrease as the film thickness is reduced. According to Puszkarski's calculation, the signal intensity and resonance-field value of surface modes is relatively insensitive to the thickness of the film until the film thickness is on the order of δ^{-1}. The resonance field value of the surface mode is expected to increase when the film thickness is reduced to a value comparable to this penetration depth. This phenomenon can be utilized to help distinguish the nature of the observed spin-wave modes, i.e., body modes or surface modes. Experimentally, the film thickness can be reduced by chemically etching the samples.

The chemical etching of the YIG films was accomplished by immersing the samples in hot (100 °C) phosphoric acid. Some of the initial work in this area of the investigation was carried out by Brown *et al.*[1] The variation of the signal intensity with film thickness for a body mode and the surface mode is shown in Fig. 12 for a film having a single surface mode. The intensity of the surface mode is seen to remain relatively unchanged when more than half of the film has been etched away, while the intensity of the first body mode has decreased by almost two orders of magnitude. As the bulk of the film is further etched the intensity of the surface mode surpasses that of the first body mode, which is the strongest mode before etching. The dashed and solid lines are model calculations to fit the experimental data. It is concluded that the single surface mode observed in the YIG films is primarily localized at the film-substrate interface. Furthermore, it is found that the penetration depth of the surface mode is on the order of 500 Å, which is a highly localized magnetic mode. The results of similar experiments on the films annealed in the laboratory are consistent with this finding in regard to this property of the surface mode.

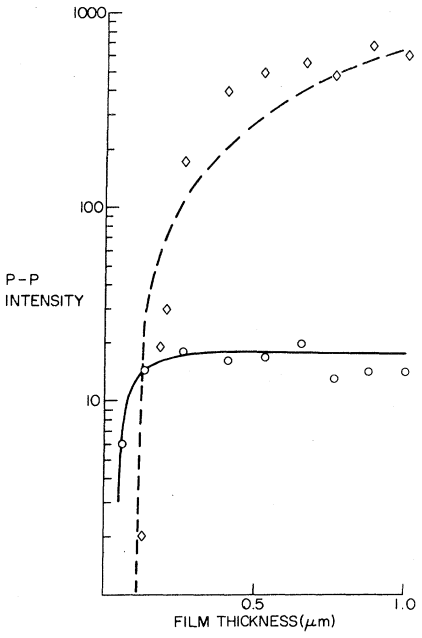

FIG. 12. Variation of the peak-to-peak intensity with film thickness for a bulk spin wave (◇) and the surface spin wave (◇). (Ref. 1.)

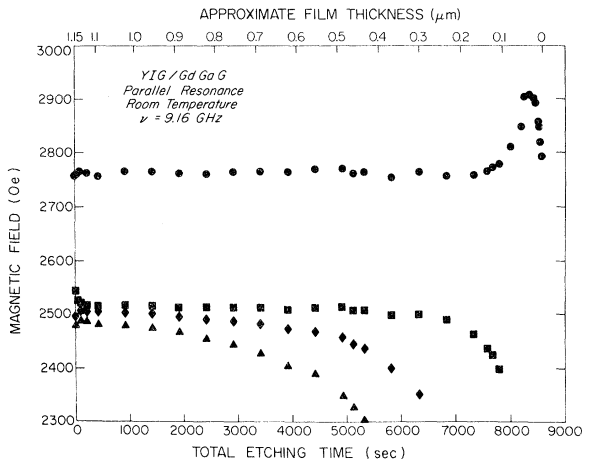

FIG. 13. Effect of etching on the resonant-field position of the high-field spin-wave modes for a YIG film having two surface modes.

Similar etching experiments have also been carried out on several YIG films showing two surface modes at the parallel resonance. A typical result is shown in Fig. 13, where the parallel resonance-field positions of the first four spin-wave modes are plotted as a function of the thickness of the film.

The film thickness was determined by assuming a constant etching rate in the bath from the initial etch until all SWR signals had disappeared. The first two high-field modes are the surface modes before etching, the third mode is the strongest and first of the body modes at parallel resonance, and the next mode is the second body mode. The field position of the high-field surface mode remains unchanged even when a large portion of the film has been etched away. The behavior of this mode near the end of the etching indicates that it is localized at the film-substrate interface. The in-

tensity of this surface mode, which is the weakest mode in the spectrum before etching, also remains unchanged as the bulk of the film is etched away, similar to the case when only a single surface mode is present.

The field position of the second surface mode decreases when approximately 100 Å of the top surface layer of the film has been etched away. The signal intensity of this mode (the second surface mode before etching) relative to the next mode (the first body mode before etching) is shown in Fig. 14. In this figure, only the second and third modes are shown; the first surface mode, which is much weaker and observed at a higher field, is not reproduced. As the top layer of the film is etched away, the intensity of the second mode increases relative to the other modes and surpasses that of the next mode when approximately 160 Å of the film is etched away. This indicates that the second mode, which is a surface mode before etching, is transformed into the first of the body modes when the top surface layer of the film is removed, while the first surface mode remains essentially unchanged. It can be inferred that the excitation amplitude of the oscillating dipole moment of the second surface mode is primarily localized at the free film surface before etching, or that the second surface mode is primarily "generated"[8] by a strong surface anisotropy field present at the top surface layer. By removing the top surface layer, the surface boundary conditions for the film becomes such that only a single surface mode can be excited in the film.

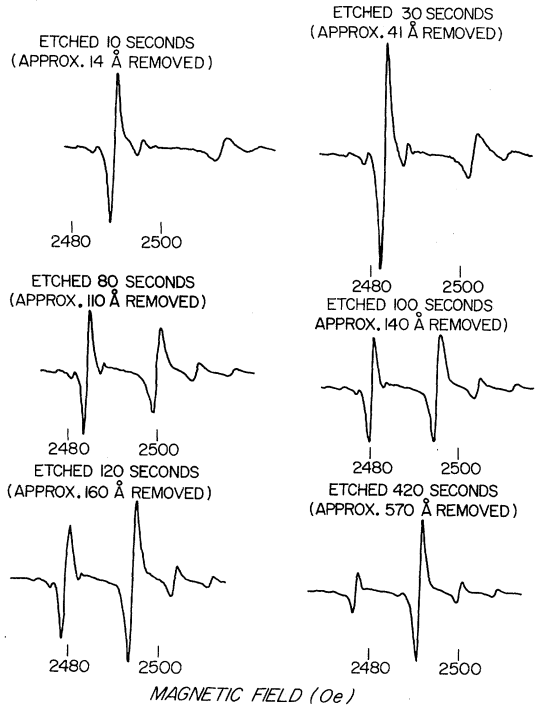

FIG. 14. Traces of the second and third high-field spin-wave modes as the film thickness is decreased by etching for a YIG film having two surface modes.

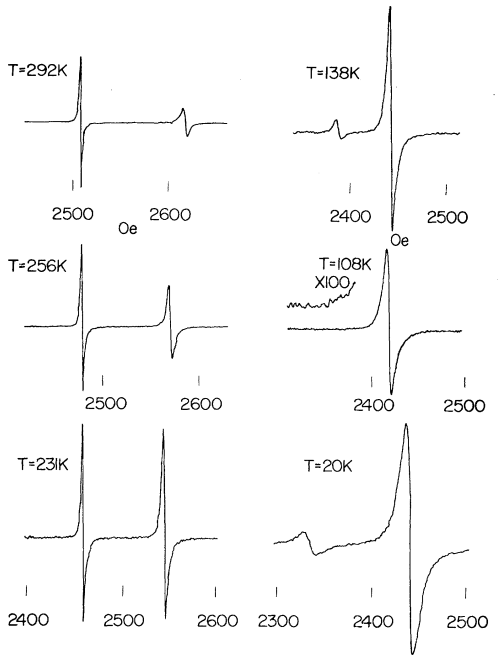

FIG. 15. Traces of the first two high-field spin-wave modes as a function of temperature for a YIG film having a single surface mode at room temperature.

In terms of a pinning model it can be deduced that by removing a thin top surface layer of the film, the source of the surface anisotropy field has been removed and the surface pinning parameter reduced to near unity, so that only one surface mode, controlled by a strong surface anisotropy at the film-substrate interface, can be excited. This further gives credence to the interpretation that the second surface mode is a quasi-antisymmetric surface mode with the excitation amplitude primarily localized at the free surface. Furthermore, this also suggests that the surface-anisotropy field at the film-substrate surface is stronger than that at the free surface. As the majority of the bulk of the film is removed, the second mode as well as the other lower field modes shift downfield, characteristic of body modes.

The first mode which remains a surface mode is observed to shift upfield as the film is reduced to a thickness of a few hundred angstroms. Meanwhile the intensity of this surface mode is observed to begin to decrease. A careful observation of the resonance signal of this surface mode also reveals

that the field position stops shifting upfield and begins to shift downfield. In fact, the surface mode becomes the only mode that can be detected when the film thickness is estimated to be only 100 Å or so. This suggests that the first surface mode, which is initially a quasisymmetric surface mode, is primarily generated by a surface anisotropy field located at the film-substrate interface.

F. Temperature dependence of the surface modes

The temperature dependence of the SWR spectrum of the YIG films has been studied from 300 to 2 K. In particular, the behavior of the surface exchange modes as a function of the sample temperature has been studied. Data have been taken for in-plane resonance, perpendicular resonance and intermediate-angle resonance. The orientation of the critical angle observed in YIG films has also been monitored as a function of the sample temperature. No significant change has been observed for the perpendicular resonance spectrum as the sample temperature is reduced, indicating that the anisotropy-field component $K_\perp$ remains relatively constant. At the parallel orientation, dramatic changes in the nature of the resonance modes and their relative intensities have been observed for samples showing surface modes in the resonance spectrum.

For a majority of the samples studied, the variation of the surface mode as a function of the sample temperature can be understood from the spectra reproduced in Fig. 15. This figure shows the first two high-field modes at parallel resonance. The weak high-field mode observed near room temperature ($T = 292$ K) has been identified as a surface mode and the next mode is the strongest and first of the body modes. As the sample temperature is reduced, the surface mode is observed to increase in intensity relative to the first body mode and become the only mode (uniform mode) observed at a temperature near $T = 109$ K. Below this critical temperature, the first mode becomes the strongest body mode observed in the spectrum. It is interesting to note that there is a strong similarity between the relative mode separations and intensities for the modes shown in Figs. 15 and 5. In both cases, the surface mode becomes the uniform mode and then transforms into the first of the body modes.

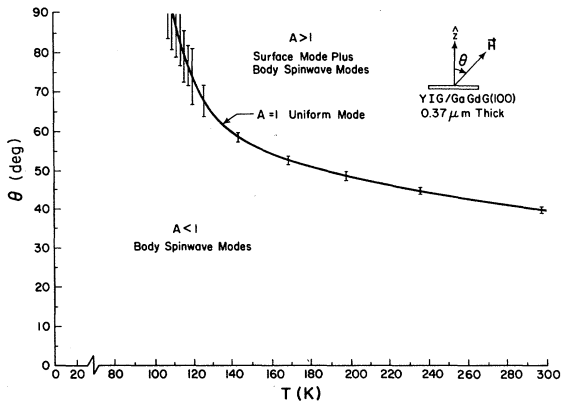

FIG. 16. Temperature dependence of the critical angle for a YIG film.

The change of the surface pinning conditions with the sample temperature is also reflected in the observed shift of the position of the critical angle with sample temperature. Figure 16 is a typical experimental curve depicting the position of the critical angle as a function of the sample temperature. The data were taken with a 0.37 μm-thick (100)-oriented film at 9.1 GHz. The critical angle θ_c is observed to increase, i.e., shift toward the parallel orientation, as the sample temperature is reduced. At a temperature near 109 K, the critical angle is observed to be located in the plane of the film. Below this critical temperature, no critical angle has been observed. It should be cautioned, however, that in some samples the surface mode is observed to disappear at a temperature near the critical temperature, instead of becoming the uniform mode.

For samples showing two surface exchange modes in the resonance spectrum, the effect of the sample temperature on the behavior of the surface mode has also been studied. Without exception, it is found that the high-field surface mode, the quasisymmetric surface mode, is observed to shift downfield and becomes degenerate with the low-field surface mode, the quasiantisymmetric surface mode. Figure 17 shows a typical example. The data points are the resonance-field separations between the two surface modes and the first body mode. At a temperature near 100 K, these two surface modes become degenerate in their resonance-field positions. Below this temperature, this surface mode is observed to appear at a nearly constant field separation above the first body modes.

IV. DISCUSSION AND CONCLUSIONS

The analysis of the magnetoexchange-branch dispersion relation in thin YIG films and the excellent agreement between theory and experiment[11] have indicated that the internal fields of these films are very homogeneous and that the spin-wave modes depend on the surface pinning conditions rather than variations in the internal field of the film. Assuming pinned surface spins, the spin-wave resonance data has been shown to fit a quadratic dispersion relation. To date nearly all ex-

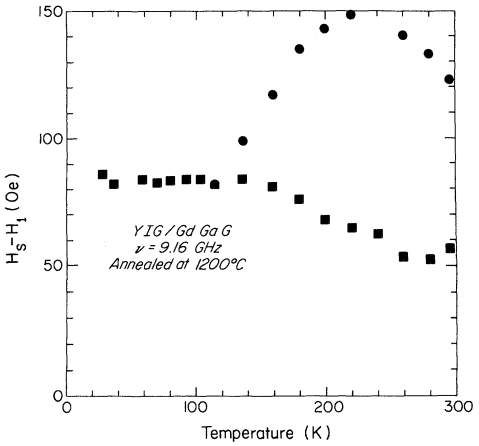

FIG. 17. Separation of the first body mode and first and second surface modes, respectively, as a function of temperature for a YIG film.

perimental results of spin-wave studies in YIG films have been limited to the perpendicular or parallel resonance geometries. As shown above, such limitations give an incomplete description of the spinwave properties.

The exchange constant D determined from perpendicular resonance spectra obtained at room temperature is found to be about (10–15)% higher than the bulk value,[11] while the value obtained from parallel-resonance data is generally slightly smaller than the bulk value. This difference along with the variation in mode separation with angle observed in this study suggests that the boundary conditions are angular dependent.

An analysis of this dependence in terms of a modified Puszkarski surface inhomogeneity model is consistent with the data if the surface spins are assumed to be subjected to a tensorial surface anisotropy.[2] For the case of a single surface mode, the in-plane and perpendicular components of the surface anisotropy associated with the free sample surface $K_{\parallel}^{f}$ and $K_{\perp}^{f}$ and the corresponding components associated with the substrate surface $K_{\parallel}^{s}$ and $K_{\perp}^{s}$ can be assigned values such that $K_{\parallel}^{f} \sim 0$, $K_{\perp}^{f} \sim 0$, $K_{\parallel}^{3} \ll 0$, and $K_{\perp}^{s} \gg 0$. This assignment of values can account for the observation of a single surface mode between parallel resonance and the critical angle where $k = 0$ and of the localization of that mode to the film substrate interface region as seen from the etching results.

The angular variation of the surface anisotropy results in the "tuning" of the wave vectors of the spin-wave modes. By varying the orientation of the applied magnetic field from parallel to perpendicular to the film surface, the highest field-mode wave vector is tuned from a pure imaginary value at parallel, to zero at the critical angle, to a real value $\sim \pi/2L$ between the critical angle and perpendicular resonance. A tunable phonon-magnon interaction dependent upon this feature has been reported.[26]

The observation of two acoustic-type surface modes is consistent with Puszkarski's theory and predictions if asymmetric boundary conditions are assumed.[27] The lack of symmetry in the boundary conditions is understandable experimentally if the environments of the two film surfaces are considered. Since all the films used are grown on GdGaG substrates, the "bottom" surface layers can be affected by the substrate. The top surface is exposed to the atmosphere. Also, a good portion of the surface-mode study involved films that were annealed. At the annealing temperatures employed, it is proposed that Gd diffuses into the YIG at the substrate surface and, in addition, at the highest annealing temperatures, a transformation occurs at the top surface. The diffusion proposed is reinforced by the observed increase in resonance-field position of the surface mode associated with the substrate surface as the annealing temperature is raised. This shift in resonance field indicates that this surface mode is becoming less and less like the uniform precession mode—that is, that it is becoming more and more localized at the film surface. This in turn agrees with the interpretation of an increased mobility of Gd into the YIG and Fe out of the YIG with increased temperature and hence an increase in the relative Gd concentration in the YIG near the substrate interface.

When the annealing temperature is 1150 °C or above, a second surface mode is observed in the spin-wave spectra. At these temperatures in the oxygen atmosphere of the annealing furnace, a surface region having a different crystal phase may be produced. The etching of several films verified the association of this second surface mode with the film's top or free surface. On removing a surface layer on the order of 100 Å the surface pinning parameter A^{F} was reduced to near unity. At the substrate surface the etching process influenced the intensity and position of the high-field surface mode before the surface region is approached. However it is estimated that the substrate surface layer can be on the order of 100 Å or less. The latter agrees with previously reported results for a single-surface-mode sample.[1]

In terms of surface anisotropies, it can be deduced that for samples in which two surface modes are observed both the parallel component of the surface anisotropy field associated with the substrate surface $K_{\parallel}^{s}$ and the parallel component associated with the free surface $K_{\parallel}^{f}$ are negative, while the perpendicular components remain positive. The increased localization of the first sur-

face mode with increased annealing temperature indicates that $K_{\parallel}^{s}$ is becoming more negative. The assignment of $K_{\parallel}^{f} < 0$ is in accord with the etching results, where the second surface mode is transformed into a body mode.

The strong temperature dependence of the surface modes can be explained in terms of a temperature dependence of the surface-anisotropy fields. The data indicates that K^{s} is large and negative at room temperature, becomes less negative as the sample temperature is decreased. The observed increase in surface-mode intensity, the decrease in the localization of the surface mode, and the shift of the critical angle toward the parallel orientation as the sample temperature is decreased are consistent with $K_{\parallel}^{s}$ decreasing in magnitude as the temperature is decreased. Below the critical temperature, where the critical angle no longer exists, $K_{\parallel}^{s}$ must become positive. Since no significant change is observed in the perpendicular spectrum as the sample temperature is changed, the $K_{\perp}^{s}$ components are significantly less temperature dependent than the $K_{\parallel}^{s}$ components.

The temperature dependence of the critical-angle results cannot be explained in terms of the volume-inhomogeneity model applicable to thin metal films.[16,20,21] The internal-field profile will have the temperature dependence of the magnetization, and as a result the changes in the critical angle predicted by the volume inhomogeneity will be very small. The large variations of the critical angle reported here and the homogeneity of the internal field as observed in the dispersion relation suggest that the pinning condition is indeed localized to the surface layers.

The magnitude of the surface-anisotropy field is observed to be of the same order of magnitude as the bulk exchange field acting between the spins. This large magnitude coupled with its unusual angle dependence suggests a possible pseudodipolar exchange interaction between the Gd ions of the substrate and Fe ions of the YIG film. The observed critical temperatures on the order of 100 °K are similar to the compensation temperature observed in mixed garnets and further suggest that the Fe-Gd interaction is responsible for the surface field. While both Fe and Gd ions are usually S-state ions which would normally be expected to have isotropic exchange interactions, the presence of the highly directional inhomogeneity in the crystal field at the interface may be responsible for the mixing of higher-order states that may give rise to the anisotropic properties.

The observation of a similar surface mode associated with the free surface after the sample is annealed at high temperature suggests the anisotropic exchange interaction may be more common than originally assumed. The lack of similarities in the temperature dependences indicates that the microscopic details of the interaction differ considerably from those of the substrate side, a result expected from the lack of symmetry between the two surfaces.

The identification of these highly localized nonpropagating surface waves in YIG films is observed to be consistent with a highly anisotropic surface interaction. The effective field produced by this interaction changes magnitude and sign with magnetic field orientation and temperature. These interactions should lead to a better understanding of the magnetic surface properties of materials.

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