sion<sup>24</sup> in Nb-Mo revealed Kohn anomalies in those concentrated alloys, but ours is perhaps the first observation on Fermi-surface effects where the mass disparity is large. Our results are also consistent with a very long-range oscillatory potential for Cu-Au and, if our comparison with Walker and Keating is more than coincidence, for  $\beta$ -CuZn as well.

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## THEORY OF SURFACE-SPIN PINNING IN FERROMAGNETIC RESONANCE

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A theory is presented which, in conjunction with previous theories, explains for the first time all notable experimental results on exchange and magnetostatic modes in thin films, including surface and bulk modes. Roughly speaking, a surface inhomogeneity thinner than  $\sim (\frac{1}{2}\Lambda)^{1/2}$  gives little pinning, while one thicker than  $\sim (\frac{1}{2}\Lambda)^{1/2}$  pins the loworder modes. The results, obtained by considering the change of  $M<sub>z</sub>$  in the surface region, reconcile the different boundary conditions used for thin-film exchange modes and for magnetostatic modes.

Since Kittel's suggestion' in 1958 that the exchange integral in ferromagnetic materials could be obtained by ferromagnetic-resonance measurements in thin films, interest in this field has increased steadily.<sup>1-5</sup> Interpretation of experi-

mental results has been obscured by a lack of understanding of the boundary conditions<sup>6</sup> at the film surfaces. The theories of Wigen and Kooi' (saturation magnetization  $M<sub>z</sub>$  of surface layer different from that of the bulk) and Portis' (parabolic  $M_{\sigma}$ ) explain the positions and critical-angle depinning, but not the intensities, of exchange modes.

A theory of surface-spin pinning<sup> $6$ </sup> is presented which explains the intensities as well as the positions in metallic and insulator films. It applies to exchange and magnetostatic modes, both bulk and surface types. Consider a saturation magnetization  $M<sub>z</sub>$  which varies across the thickness of the film as illustrated in Fig.  $1.^7$  In the surface region of thickness  $\epsilon$ ,  $M_z$  drops from its bulk value to zero. In the bulk region  $M<sub>z</sub>$  is some function of z' such as<sup>3</sup>  $M_0-(2z'/S)^2\tilde{\Delta M}$ .

In  $\perp$  resonance,<sup>6</sup> the ferromagnetic normal modes having exchange energy  $\leq 2\pi\hbar |\gamma| \Delta M$  are pinned in the bulk region according to the theory of Portis.<sup>3</sup> For  $\epsilon = 0$  and  $\Delta M = 0$  the modes are unpinned' since all spins must precess at the same frequency in a normal mode and a nonzero slope would give an extra torque on the surface spins because of the missing neighbors. For  $\Delta M$  $=0$  and  $\epsilon \neq 0$ , the modes will remain unpinned for  $\epsilon^2 \ll \epsilon_{cr}^2$ , where  $\epsilon_{cr}^2 \approx \frac{1}{2}\Lambda$  and  $\Lambda$  is the exchange constant. For "80-20" Permalloy,  $\sqrt{\Lambda} = 160 \text{ Å}$ , and for yttrium iron garnet (YIG),  $\sqrt{\Lambda} = 568$  Å.

If  $\epsilon^2 \ll \epsilon_{\rm cr}^2$  is not satisfied, the surface exchange cannot hold  $d\vec{m}/dz' = 0$  at the surface. Since the spins in the surface region are "off resonance" when the spins in the bulk are "on resonance" and the surface spins are exchange and dipole coupled to the bulk spins, the low-order modes are pinned. However, the higher order modes are only partially pinned, as will be shown.

Consider the equation of motion  $d\vec{M}/dt = -|\gamma|\vec{M}|$  $\times$ H, where the total effective field is  $H = H_i + \Lambda \nabla^2 \vec{M}$ + $\bar{h}_d$ , where  $\bar{H}_{i}$ + $\bar{H}_{app}$  – $\hat{z}'$ 4 $\pi M_z$  cos $\theta_m$ ,  $\bar{H}_{app}$  is the applied field,  $\overline{h}_d = \frac{\overline{z}^T 4\pi M_X \sin \theta_m}{\overline{w}}$  is the microwave demagnetization field, and  $\theta_m$  and the coordinate axes are shown in Fig. 2. With  $\overline{M} = \hat{z}M_z + \overline{m}$  and with  $exp(i\omega t)$  time dependence, the linearized equations of motion of  $m_x$  and  $m_y$  can be diagona-



FIG. 1. Variation of the saturation magnetization  $M_z$  across the thickness (z' axis) of the film.

lized to give  $\Lambda M_z d^2m/dz^2 + \kappa_+^2m = 0$ , where m is the linear combination of  $m<sub>x</sub>$  and  $m<sub>y</sub>$  (e.g.,  $m = m<sub>x</sub>$ )  $+im_{\nu}$  in the circular precession approximation<sup>8</sup>) obtained from the diagonalization and

$$
\kappa_{+}^{2} = [(2\pi M_{z} \sin^{2}\theta_{m})^{2} + \omega^{2}/\gamma^{2}]^{1/2}
$$

$$
-H_{i} - 2\pi M_{z} \sin^{2}\theta_{m} - \Lambda d^{2} M_{z}/dz'^{2}. (1)
$$

In the surface region, the term  $\Lambda d^2M_z/dz^2$  is of the order of  $D(\pi/\epsilon^2)$ . First consider the sharpboundary case in which  $D(\pi/\epsilon^2)$  is much larger than the sum of the other terms on the right-hand side of (1). For perpendicular resonance this means  $D(\pi/\epsilon^2) \gg 2\pi M_0 + (\Delta \omega_{\perp}/|\gamma|)$ , or

$$
\epsilon^2 \ll \epsilon_{\text{CFL}}^2, \qquad \qquad \text{CFL}
$$
\n
$$
\epsilon_{\text{CFL}} = (\frac{1}{2}\Lambda)^{1/2} \left[ 1 + \Delta \omega_{\perp} / 2\pi |\gamma| M_0 \right]^{-1/2}, \qquad (2)
$$

where  $\Delta \omega_{\perp} = \omega - |\gamma| (H_{app} - 4\pi M_0) \approx |\gamma| D k_n^2$  for the nth exchange mode. Since modes with  $Dk_n^2 \gg 2\pi M_0$ are seldom observed in practice, (2) gives  $\epsilon_{\text{crl}}$  $\approx (\frac{1}{2}\Lambda)^{1/2}$ . When (2) is satisfied, the equation above (1) for *m* reduces to  $M_z d^2m/dz'^2 = md^2M_z/$  $dz^2$  in the surface region. Integrating once with  $m = M_z = 0$  at  $z' = \pm \frac{1}{2}S$  gives  $M_z dm/dz' = mdM_z/dz'$ . Since *m* and  $M_z$  are continuous at  $z' = \pm(\frac{1}{2}S - \epsilon)$ , the equation above  $(1)$  for  $m$  can be solved in the bulk region  $|z'| < \frac{1}{2}S - \epsilon$  with the boundary condition  $(1/m)dm/dz' = (1/M_z)dM_z/dz'$  at  $z' = \pm(\frac{1}{2}S-\epsilon)$ .

Thus the effect of the sharp surface region is to establish this boundary condition for the bulk region. This result can also be obtained quite simply by requiring that the net additional torque on a surface spin caused by the missing neighboring spins be equal to zero for a perfectly sharp



FIG. 2. Schematic illustration showing fields, angles, and coordiante systems used in the text. The axis of quantization is  $z$ , and the  $z'$  axis is normal to the film surface.

boundary  $(\epsilon = 0)$ . The equation-of-motion method shows that the result is also valid for  $\epsilon \neq 0$  as long as (2) is satisfied. It is not difficult to show<sup>9,10</sup> that this boundary condition gives only a small amount of pinning for the small values of  $\Delta M$  observed to date.<sup>4</sup>

For  $\epsilon^2 \ll \epsilon_{cr}^2$  not satisfied, which we call the thick-boundary case, consider the simple step model of  $M_z = M_B$  in the bulk region and  $M_z = M_S$ in the surface regions, as illustrated by the dashed lines in Fig. 1. Since the gross features of the pinning do not depend strongly on the shape of  $M_z$  in the surface region, this model is appropriate for a smoothly decreasing  $M_z$  of thickness  $\epsilon$ , as well as for a sharp surface layer of thickness  $\frac{1}{2}\epsilon$ .

It can be shown from the equation of motion or by considering the torques on the spins at the sharp boundary at  $z' = -\frac{1}{2}(S-\epsilon)$  between  $M_S$  and  $M_B$  that  $m_B'/m_B = (M_S/M_B)^2 (M_S'/M_S)$ , where the prime denotes derivative with respect to  $z'$  and B and S denote

$$
z' = \lim_{\delta \to 0} \left[ -\frac{1}{2}(S - \epsilon) \pm \delta \right],
$$

respectively. Solving for  $m$  in  $\perp$  resonance with this continuity condition and with  $M_S < M_B$  gives  $m = m_0 \cos k_n z'$ , where  $k_n$  are the roots of tanp  $=\frac{1}{2}(M_S/M_B)^2|k_{+S}|S\tanh(\frac{1}{2}|k_{+S}|\epsilon)/p$ . Here  $p=\frac{1}{2}k_{-S}S$ and  $k_{+S}^2 \cong [4\pi (M_B-M_S)-Dk_B^2]/\Lambda M_S$  is the magnitude of  $\kappa_{+}^{2}/\Lambda M_{z}$  from (1) in the surface region. From this result with  $\epsilon \, {\geq} \, 2 / |k_{+S}|, \; M_S \! = \! \frac{1}{2} M_B, \;$  and  $Dk_n^2 \leq 2\pi M_B$ , it is easy to show that

$$
I_n = \frac{8}{\pi^2} \frac{1}{n^2} \text{ for } n < n_{2-4} = \frac{1}{2\sqrt{\pi}} \frac{S}{\sqrt{\Lambda}} + \frac{1}{2};\tag{3}
$$

$$
I_n = \frac{2}{\pi^3} G \tanh^2 \left[ \left( \frac{\pi G}{n} \right)^{1/2} \epsilon \right] \frac{1}{(n-1)^4}, \quad n > n_{2-4}; \quad (4)
$$

$$
k_n = (n-p_n)\pi/S, \quad n = 1, 2, 3, \cdots, \quad 0 < p_n < 1,\tag{5}
$$

where  $G = |1 - Dk_n^2 / 2\pi M_0|$ . The present pinning mechanism gives the same value of critical angle for depinning as predicted by the previous theofluxtion tor depinding as predicted by the previous theo-<br>ry<sup>5</sup>; the spins are unpinned when  $k_{+} \equiv \kappa_{+}/\Lambda M_{Z}$  has the same value in the bulk and surface regions.

For  $\perp$  resonance, (3) and (4) are the central results. For very high-order modes, the  $Dk_n^2$  term in G dominates and  $I_n \sim (n-1)^{-8}$ ; otherwise (4) gives  $I_n \sim (n-1)^{-4}$ . The value np of n below which the modes are pinned by the Portis mechanism is determined by the relation  $Dk_n^2 \cong 2\pi\Delta M$ , with  $k_n$ given by (5). If the linewidths  $\Delta H_n$  are not the same for all modes, the values of  $I_n$  given in (3) and (4) [which are normalized as  $I_n = S^{-1} \int dz m(z) |z|$  with  $\int dz m(z)^2 = 1$  must be multiplied by  $1/\Delta H_n$ .

The experimental intensities and spacing of the  $\perp$  resonance lines in the classic sample PK-23 of Kooi, Wigen, and co-workers' agree well with the theoretical results (3) and (4). For this sample,  $n_{2-4} \approx 13$  and  $np \approx 10$ . The first five even modes, which have  $n < np$ , have large intensities and nearly linear spacings, in agreement with Portis's theory.<sup>3</sup> For the modes with  $n \ge 13$  (so that  $n \geq n_{2-4}$  is satisfied) the experimental (in square brackets) and normalized theoretical (in parentheses) values  $I_n \sim (n-1)^{-4}$  are as follows: , [3.50], (3.71);  $I_{15}$ , [1.90], (1.86);  $I_{17}$ , [1], (1);  $I_{19}$ , [0.60], (0.56);  $I_{21}$ , [0.34], (0.32);  $I_{23}$ , [0.17], (0.18). The orders of magnitude are also correct:  $I_{23}/I_1$ , [(2-3)×10<sup>-3</sup>], (1.4×10<sup>-3</sup>).

The agreement with the theory for the thickboundary case is excellent. Nisenoff and Terhune' also found that their data for Permalloy films could be explained by a surface layer (corresponding to the thick-boundary case in the present theory). These results and others<sup>5,1</sup> are reasonable for metallic films, which are well known to have quite inhomogeneous  $M_z$  in general; there should be sufficient rounding of  $M_z$  at the surfaces such that  $\epsilon^2 \ll \epsilon_{cr}^2 \approx (110 \text{ Å})^2 \text{ is } \text{not}$ satisfied. This appears to be the case in all metallic-film results in the literature with only one tallic-film results in the literature with only<br>exception,  $11$  where  $M_Z$  was purposely made as nearly constant as possible.

For extremely homogeneous films it is apparently possible to have  $\epsilon^2 \ll \epsilon_{c}r^2$  so that there is very little pinning. Waksmann, Massenet, Escudier, and Kooi<sup>11</sup> found that for expitaxial  $81\%$ -Ni, 19%-Fe films deposited on silver on freshly cleaved mica at 330°C and less than  $10^{-6}$  Torr, the intensities of all higher order modes were less than  $10^{-3}$  times the intensity of the principa mode. For single-crystal YIG thin films grown mode. For single-crystal YIG thin films grows<br>by Mee and co-workers,<sup>10</sup>  $M_z$  is expected to be much more homogeneous than in metallic films, and  $\epsilon_{cr}^2$  is ~13 times larger than in Permalloy. Thus the theory predicts very little pinning at the large surfaces of these films, as verified by preliminary measurements  $(I_3/I_1 \approx 0.7 \times 10^{-3})$ .<sup>10</sup>

For  $\parallel$  resonance, the central experimental result for a large number of measurements in metallic films is that fewer lines are observed in  $\parallel$ resonance than in  $\perp$  resonance, there being typically two or three strong lines in  $\parallel$  resonance.<sup>4</sup> There are several reasons for this, the most important one being that  $n_{2-4}$  is usually less than 4. For example, for  $S = 0.3 \mu$  and  $\sqrt{\Lambda} = 160 \text{ Å}$ ,  $(n_{2-4})_{\parallel} \approx 2.7$ . Thus, only the first two even modes  $(n$ 

 $=1,3$ ) are strongly excited. Second, the potential well is "negative"<sup>4</sup> in  $\parallel$  resonance; thus there is no Portis-type pinning.<sup>3</sup> Third,  $\epsilon_{cr}^2$  is larger in If resonance than in  $\perp$  resonance (typically ~6 times larger in Permalloy films). Fourth, the intensities are smaller in  $\parallel$  resonance than in  $\perp$ intensities are smaller in  $\|$  resonance than in<br>resonance even when  $\epsilon^2 \ll \epsilon_{\rm cr}^2$ <sup>2.10</sup> Finally, if a two-magnon process controls the linewidth  $\Delta H$ , then  $\Delta H$  is usually larger in  $\parallel$  resonance than in  $\perp$  resonance. This last factor should be more important in YIG films than in metallic films.

The pinning at the large surfaces of singlecrystal YIG films is small for the exchange and magnetostatic bulk and surface modes, as discussed above. The spins within a distance  $\sim$ S from the small edges of the film have internal fields and microwave demagnetization fields which are different from those of the bulk spins. This pins the edge spins in much the same manner as does a surface layer of  $M_S$ .<sup>7</sup> The results are<sup>10</sup> that the modes with  $\Delta\omega < 3\pi|\gamma|M_0|$  where  $\Delta\omega$  $\equiv \omega - \gamma H_i$  for  $\perp$  resonance and  $\Delta \omega = \omega - \gamma H_{app}$  $\times (H_{\text{app}} + 4\pi M_0)^{1/2}$  for  $\parallel$  resonance } are pinned and those with  $\Delta \omega > 3\pi |\gamma| M_0$  have  $I_n \sim 1/n^4$ . Since the largest value of  $\Delta\omega/|\gamma|$  observed to date is  $\approx$  500 Oe, and  $3\pi M_0 \approx$  1310 Oe in YIG at room temperature, the theory predicts that all these modes should be pinned at the film edges, in agreemen<br>with experiments.<sup>10</sup> with experiments.

Since the experiments and theory indicate that  $dm/dz \approx 0$  at the large surfaces of the YIG films, it would appear that the Damon and Eshbach<sup>12</sup> (DE) surface modes, which have nonzero slopes at these surfaces, would not exist. Fortunately this is not the case. The slope of  $m$  is small at the surface, but  $m$  bends down and rapidly approaches the exponentially decaying  $m$  of DE.<sup>13</sup> In particular,  $m_{\text{DE}} \sim \exp(-k_x x)$  is a good approximate solution as long as  $Dk_x^2 \ll \omega/|\gamma|$ .<sup>10</sup> For YIG mate solution as long as  $Dk_{\chi}{}^2 \!\ll\! \omega/|\gamma|.^{\bf 10}~$  For YIG at X band in  $\parallel$  resonance with  $k_z = 0$  (so that  $k_y$  $= \pm k_{\chi}$ , the DE solutions should be valid for  $|k_{\chi}|$  $56\times10^4$  cm<sup>-1</sup>. This result is consistent with the observations of surface waves with  $|k_y| = 0$ -700 cm<sup>-1</sup> in thin single-crystal YIG films.<sup>14</sup>  $\text{cm}^{-1}$  in thin single-crystal YIG films.

The present theory affects the interpretation of experiments to determine  $D$  and the spin-wave dispersion relation: Phillips 's' assumption of temperature-independent pinning is supported by the theory. Kooi, Shanabarger, and Wigen<sup>15</sup> showed that the spacing of the Portis-pinned modes should decrease as  $T^{3/2}$  and suggested that the same might be true for the modes pinned at the sample surface. The present theory indicates that this is not the case. The temperature

independence of the roots  $k_n = n\pi/S$  of the tan $p$ equation for  $n \leq n_{2-4}$  shows that the pinning of these modes is independent of temperature. Indeed, the  $n^2$  spacings and  $1/n^2$  amplitudes indicate that the modes are pinned at the surfaces. The theoretical values of  $n_{2-4} \approx 5$  and 9 for S  $=1800$  and 4200 Å, respectively, are consistent with  $I_n \sim 1/n^2$ .

The experimental and theoretical agreement of the mode intensities of the beautiful experimental data of Weber and Tannenwald<sup>1</sup> is excellent:  $I_{17}$ ,  $[2.25]$ ,  $(2.33)$ ;  $I_{19}$ ,  $[1.5]$ ,  $(1.49)$ ;  $I_{21}$ ,  $[1]$ ,  $(1)$ ;  $I_{23}$ , [0.80], (0.70);  $I_{25}$ , [0.49], (0.50);  $I_{27}$ , [0.32],  $(0.36)$ ;  $I_{29}$ ,  $[0.125]$ ,  $(0.125)$ ;  $I_{31}$ ,  $[0.09]$ ,  $(0.073)$ ;  $I_{33}$ , [data missing];  $I_{35}$ , [0.28], (0.28);  $I_{37}$ , [0.016],  $(0.018); I_{39}, [0.011], (0.012); I_{41}, [0.0071], (0.0078);$  $I_{43}$ , [0.0037], (0.0053). The theoretical values, which are normalized at  $I_{21}$  and  $I_{29}$ , are  $I_n \sim (n-1)$ for  $n = 17-27$  (for  $n > n_{2-4}$  and  $\Delta \omega_1 / |\gamma| \approx D k_n^2 < 2 \pi M$ <br>  $\approx 5 \text{kOe}$ ) and  $I_n \sim (n-1)^{-8}$  for  $n = 29-43$  (for  $D k_n^2$  $>2\pi M_0$ ).

The deviation from linearity of  $ln(H_n-H_0)$  vs inn, which Weber and Tannenwald attributed to a long-range exchange interaction, is eliminated when the pinning of the present theory is used. The modes with  $n \ge 15$  are nearly unpinned and have  $k_n \approx (n-1)\pi/S$ , rather than  $n\pi/S$ . The experimental intensities verify that the modes cannot be pinned, as previously assumed, since  $I_n \sim (n-1)^{-4}$ and  $(n-1)^{-8}$  rather than  $1/n^2$ . Replotting the points in their Fig. 1 one unit to the left entirely eliminates the deviation of  $\ln(H_n - H_0)$  vs  $\ln n$  from linearity, thus removing the disagreement with the neutron results of Menzinger and co-work<br>ers.<sup>16</sup> ers.

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