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Observation of Spin Wave Resonance in Ni Thin Films after Adsorption of Oxygen*

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Ni thin films were deposited in ultrahigh vacuum (UHV) onto soft glass substrates. Film thicknesses ranged from 74 to 837 Å. On films in UHV, only the uniform precession mode was observed during the microwave experiments. Admission of gases such as H₂, H₂O, O₂, N₂O, and air caused a lowering of the resonance field of the uniform precession mode. This effect was interpreted earlier as a relief of compressive stress related to surface tension. After adsorption of gases the films were virtually stress free. In the case of O₂, N₂O, and air admission a standing spin wave resonance (SWR) mode (p = 1) slowly developed in films thicker than 400 Å after the lowering of the resonance field of the uniform precession mode was observed. From the fact that in UHV and in the presence of H₂ and H₂O no SWR was found, while development of the first mode was slow in oxygen-containing atmosphere and was thought to be related to the growth of a NiO layer, it may be concluded that spin wave excitation is made possible in this case as a result of surface spin pinning due to ferromagnetic-antiferromagnetic exchange coupling (Meiklejohn and Bean) rather than surface anisotropy (Néel). The surface spin pinning was found to be weak but the intensity of the first mode increased rapidly with film thicknesses above 400 Å. On the basis of the surface spin pinning model proposed by Kittel the exchange constant A was assigned an average value of 0.74×10⁻⁶ erg/cm.

INTRODUCTION

The vacuum system in which the films were prepared and measured has been described elsewhere.1 Ni thin films (area 8×30 mm) were evaporated onto extremely well-degassed soft glass substrates (Corning 0211, alkali borosilicate, thickness 0.3 mm) at 25–35°C. The vacuum during evaporation was in the 10⁻⁷ Torr range, deposition rates varied from 40 to 100 Å/min. The nickel source consisted of a well-degassed W wire coated with Ni (99.995%), mounted approximately 80 mm from the substrate and parallel to it. Using this arrangement, the maximum film thickness reached was 837 Å. After evaporation the vacuum dropped to values below 2×10⁻¹⁰ Torr. It is believed that no appreciable gas adsorption takes place under these conditions. Ferromagnetic resonance spectra of the films in UHV were taken as soon as the films were prepared. During measurements the film plane was perpendicular to the static magnetic field. The effects of gas adsorption on the resonance spectra were studied by admission of gases to the system via a fine control valve. During all experiments the films were at room temperature. H₂ and O₂ (impurities less than 2 ppm) and air were dried via a liquid-nitrogen trap; H₂O and N₂O were cleaned by repeated freezing and pumping.

In a previous study1,2 films prepared and measured in UHV were found to be in a state of compressive intrinsic stress. This was concluded from the fact that the values of the resonance fields (HR) were more than 9200 G. Adsorption of O₂, N₂O, H₂, H₂O, and air caused stress relief. [For a stress-free film the resonance equation of the uniform precession mode is w/γ=H_{RL}. Taking HR=4900 emu/cm³, w=2πν (p=9.3×10⁹ sec⁻¹) and γ=2.80·10⁵ (γN=2.18) yields H_{RL}=9200 G.] These observations were explained by means of the surface tension and a simple model was proposed to which the observed resonance fields agreed quantitatively. Clearly separated from the stress relief effect was the appearance of a spin wave resonance mode in the spectra of films thicker than 400 Å in atmospheres containing O₂ and N₂O; the latter effect is reported in detail in this paper.

SWR in Ni thin films was earlier observed by Kimura and Nose.3 Their films were prepared on unheated glass substrates under vacuum conditions of 4×10⁻⁵ Torr; observations were made in air. A 2000-Å thickness film showed 4 SWR peaks. The average value of the exchange coupling constant A was calculated to be 0.75×10⁻⁶ erg/cm.

Lykken et al.4 found SWR in permalloy thin films in the vacuum in which the films had been prepared (approximately 1×10⁻⁷ Torr) as well as in air. They concluded that spin pinning exists in a modest vacuum.

EXPERIMENTAL RESULTS

The results obtained on 16 films ranging in thickness from 74 to 837 Å are summarized in Table I. Films as prepared and measured in UHV show decreasing resonance fields with increasing film thickness, in accordance with the surface tension model that was mentioned before. In the resonance spectra of these ultraclean films only the uniform precession mode

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TABLE I. Experimental results.

<table>
<thead>
<tr>
<th>Film thickness (Å)</th>
<th>$H_{p=1}$ (not observed)</th>
<th>$H_{p=1}$ (not observed)</th>
<th>After exposure to O$_2$, air, or N$_2$O (atmospheric pressure, long time)</th>
</tr>
</thead>
<tbody>
<tr>
<td>74</td>
<td>10 400</td>
<td>8870</td>
<td>$H_{p=1}$ $H_{p=1} H_{p=1} (t)$ $\Delta H_{p=1}$ $\Delta H_{p=1}$ $\Delta$</td>
</tr>
<tr>
<td>113</td>
<td>10 270</td>
<td>8910</td>
<td>220</td>
</tr>
<tr>
<td>118</td>
<td>10 150</td>
<td>8910</td>
<td>220</td>
</tr>
<tr>
<td>156</td>
<td>9 880</td>
<td>9150</td>
<td>220</td>
</tr>
<tr>
<td>204</td>
<td>9 780</td>
<td>9240</td>
<td>140</td>
</tr>
<tr>
<td>267</td>
<td>9 600</td>
<td>9260</td>
<td>160</td>
</tr>
<tr>
<td>402</td>
<td>9 420</td>
<td>9160</td>
<td>7220 $H_{p=1}$ 1940 $0.019$ 210 $0.78$</td>
</tr>
<tr>
<td>445</td>
<td>9 440</td>
<td>9180</td>
<td>7860 $H_{p=1}$ 1320 $0.031$ 200 $0.65$</td>
</tr>
<tr>
<td>462</td>
<td>9 380</td>
<td>9260</td>
<td>8780 $H_{p=1}$ 1320 $0.020$ 190 $0.75$</td>
</tr>
<tr>
<td>477</td>
<td>9 470</td>
<td>9260</td>
<td>8780 $H_{p=1}$ 9200 $0.018$ 220 $0.67$</td>
</tr>
<tr>
<td>524</td>
<td>9 510</td>
<td>9260</td>
<td>8140 $H_{p=1}$ 1120 $0.046$ 150 $0.77$</td>
</tr>
<tr>
<td>550</td>
<td>9 510</td>
<td>9260</td>
<td>8250 $H_{p=1}$ 1010 $0.052$ 150 $0.76$</td>
</tr>
<tr>
<td>574</td>
<td>9 400</td>
<td>9260</td>
<td>8390 $H_{p=1}$ 870 $0.070$ 210 $0.71$</td>
</tr>
<tr>
<td>657</td>
<td>9 380</td>
<td>9260</td>
<td>8530 $H_{p=1}$ 700 $0.090$ 190 $0.75$</td>
</tr>
<tr>
<td>808</td>
<td>9 350</td>
<td>9260</td>
<td>8760 $H_{p=1}$ 490 $0.245$ 150 $0.79$</td>
</tr>
<tr>
<td>837</td>
<td>9 400</td>
<td>9260</td>
<td>8780 $H_{p=1}$ 460 $0.230$ 160 $0.80$</td>
</tr>
</tbody>
</table>

* $H_p$ = resonance field ($H_{p=m}$) in G. 

† Linewidth, defined as the field separation in gauss between the inflection points of the absorption curve. 

$\Delta H_{p=1}$ Exchange constant in $10^{-6}$ erg/cm. average value $0.74 \times 10^{-6}$ erg/cm.

($p=0$) was found. After exposure to H$_2$ (477 and 524 Å films) and H$_2$O (550 and 837 Å films) the resonance fields shifted towards lower values (stress relief) but no SWR modes appeared. When O$_2$ was admitted to these films the resonance fields showed a small additional shift and a SWR mode (labeled $p=1$) slowly developed. Labeling $p=1$ of the appearing SWR mode is justified in view of the fact that the surface spin pinning model is accepted for the interpretation of the results of this study. In this model the $p=1$ mode will be the strongest when spin pinning exists at both interfaces. From earlier work$^1$ it can be concluded that oxidation takes place at both the film/gas and film/substrate interfaces, while this study shows

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**Fig. 1.** Examples of ferromagnetic resonance spectra. (a) Development of weak spin wave mode (relatively thin film); (b) development of stronger spin wave mode (thicker film).
that oxidation leads to surface spin pinning. The resonance fields after exposure to oxygen of all but the 74 and 113 Å thickness films were 9200 ± 60 G, indicating that these films were practically stress free. The two thinnest films exhibited lower resonance fields after exposure to oxygen of all but N\textsubscript{2}O, and air was noticed with all films thicker than 200 Å. The development of a SWR mode in continuity of the films. The relatively large linewidths indicating that these films were practically stress free.

The latter development could be observed at pressures as low as 1×10\textsuperscript{-4} Torr. The resonance spectra are illustrated in Fig. 1 for 445- and 837-Å thickness films. The relative intensity of the spin wave mode \( p=1 \) increased slowly with exposure time to oxygen (see Table II). (For the intensity \( I \), the height of the signal in derivative representation was taken.) The values of \( I_{p=1}/I_{p=0} \) after very long exposure time to air are plotted in Fig. 2 as a function of film thickness. The spacings of the modes \( p=0 \) and \( p=1 \) increased with decreasing film thickness, in accordance with the resonance condition for the surface spin pinning model

\[
\omega/\gamma = H_{R1} - 4\pi M_e + (2A/M_e)k^2, \quad (1)
\]

where \( A \) is the exchange coupling constant, \( k \) is the wave vector which is equal to \( p\pi/t \) in which \( t \) is the film thickness and \( p \) is the mode number. While the SWR modes were still developing, the spacings of the modes \( p=1 \) and \( p=0 \) were observed to be constant. Using Eq. (1) the exchange constant \( A \) was calculated for all films; the average value for \( A \) was found to be 0.74×10\textsuperscript{-6} erg/cm, which is in excellent agreement with the value reported by Kimura and Nose.

**DISCUSSION AND CONCLUSIONS**

For the interpretation of spin wave resonance in magnetic thin films, two different models are being used, the surface spin pinning model and the volume inhomogeneity model. As indicated before, the surface spin pinning model proposed by Kittel has been adopted to explain the results of his study. The main reason for this is that surface effects, which are believed to play the most important role in this work, are the key to the interpretation via the surface spin pinning model. Using the surface spin pinning model, there is no need for assuming a nonuniform distribution of the magnetization normal to the film plane. Such an assumption is a requirement for the volume inhomogeneity model but is unlikely to be valid for films prepared in UHV onto well-outgassed substrates as used in this study.

As pinning mechanisms for the surface spin pinning model the surface anisotropy and exchange anisotropy have been proposed. Both mechanisms are based on the anisotropic environment of the surface atoms. Surface anisotropy is present at clean or contaminated surfaces while exchange anisotropy exists as a result of the presence of antiferromagnetic material on the surface of ferromagnetic material (for example, NiO on Ni). It appears that on the basis of this study a distinction between the two mechanisms becomes evident: From the fact that clean films in UHV do not show SWR modes, it may be concluded that the surface anisotropy cannot be responsible for the excitation of SWR and does not lead to surface spin pinning. Changes in the surface anisotropy, as will occur on chemisorp-

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**Table II. Effect of exposure time to oxygen on the relative intensity of the spin wave resonance mode \( p=1 \).**

<table>
<thead>
<tr>
<th>Film thickness (( t ) (Å))</th>
<th>10 min</th>
<th>20 min</th>
<th>30 min</th>
<th>40 min</th>
<th>50 min</th>
<th>( \infty ) time</th>
</tr>
</thead>
<tbody>
<tr>
<td>445</td>
<td>( 0^a )</td>
<td>0.007\textsuperscript{*}</td>
<td>0.013\textsuperscript{*}</td>
<td>0.017\textsuperscript{*}</td>
<td>0.018\textsuperscript{*}</td>
<td>0.031\textsuperscript{d}</td>
</tr>
<tr>
<td>524</td>
<td>0.009\textsuperscript{b}</td>
<td>0.014</td>
<td>0.014</td>
<td>0.016</td>
<td>0.026</td>
<td>0.046\textsuperscript{d}</td>
</tr>
<tr>
<td>837</td>
<td>( 0^b )</td>
<td>0.010</td>
<td>0.014</td>
<td>0.014</td>
<td>0.014</td>
<td>0.230\textsuperscript{d}</td>
</tr>
</tbody>
</table>

\( ^a \) First exposed to H\textsubscript{2}O, next to air.

\( ^b \) First exposed to H\textsubscript{2}S, next to air.

\( ^{\text{d}} \) In 1×10\textsuperscript{-4} Torr O\textsubscript{2}.
tion of H₂ or H₂O₂ and formation of a nickel hydride layer (diamagnetic) or adsorbed H₂O layer does not induce SWR either. Kooi et al.⁴ found that permalloy films reduced in H₂ show indeed weak pinning at the reduced surface. From the results of this study it is clear that SWR does occur after oxidation (O₂, air, N₂O), at room temperature. The appearance of SWR is a slow process, indicating that build-up of an antiferromagnetic NiO layer (10 Å or more) is needed to achieve sufficient pinning. Presence of only a monolayer of oxygen does not lead to measurable effects.⁸ From

This it may be concluded that the ferromagnetic-antiferromagnetic exchange anisotropy only leads to surface spin pinning and excitation of spin wave resonance in this case. The surface spin pinning is relatively weak as may be seen from the fact that in films thinner than 400 Å no SWR was found, while the intensity of the appearing SWR mode for films thicker than 400 Å increases rapidly with film thickness (Fig. 2).

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