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Magnons in superlattices: A light scattering study

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We have performed a study of magnons in Mo/Ni metallic superlattices with the use of Brillouin scattering. The magnetic field and modulation wavelength dependence of the magnon frequencies are found to be in reasonable agreement with theoretical predictions. Certain features in the spectra which are characteristic of a superlattice and cannot be expalined by a trivial superposition of the spectra of the individual constituent layers are presented and discussed.

I. INTRODUCTION

In recent years there has been considerable interest in the class of materials referred to as superlattices. These materials have been shown to possess properties which differ from those of their constituents. Investigations to date have included electronic band structures, elastic behavior, superconducting properties, electrical resistivity, and structure determination.[1] Studies of the magnetic behavior of superlattices have consisted of the determination of the static magnetization,[2,3] neutron scattering,[4] and ferromagnetic resonance.[5] All of these results can be explained as being due to simple thin-film effects. The existence of a superlattice, however, is expected to greatly modify the magnon spectra of these materials as has been shown in two recent publications. Grünberg and Mika[6] calculated the magnon energies for a propagation direction parallel to the surface of the superlattice and perpendicular to the magnetization, and presented an experimental spectrum to support their calculations. Camley, Rahman, and Mills[7] have performed calculations for general propagation directions and calculated the coupling of the various modes to be expected in a light scattering study. In a recent paper[8] we have reported the experimental observation of certain predictions of these theories.[6,7]

In this paper we present the results of an extensive light scattering study of the magnon spectra of Mo/Ni superlattices. Our results agree qualitatively with the predictions of theory. Reasons for the observed discrepancies are discussed and it is concluded, within the accuracy of our experiments, that the theory is capable of explaining all observations.

II. THEORY

Theoretical studies of magnons in superlattices, in which the exchange interaction has been neglected, are presented in Refs. 6 and 7. Here we reproduce their results for the special case where the applied field (and hence the magnetization) is perpendicular to the propagation direction of the magnon, and lies in the plane of the sample.

Two types of modes are discussed: (a) modes arising from the interaction of surfacelike magnons in each magnetic layer, and (b) modes arising from the interaction of standing spin waves in each layer.

It is found, for the former, that a band of excitations exists in the range of frequencies given by

$$\nu=\gamma[H(H+4\pi M)+4\pi^2M^2w]^{1/2} . \tag{1}$$

Here γ is the gyromagnetic ratio, H is the applied magnetic field, M is the magnetization of the individual magnetic layers, and w is a number that depends on the thicknessses of layers and on the wave vector of the magnon. From the results of Ref. 7 it is straightforward to show that

$$w=\frac{2\sinh(d_1Q_{\|})\sinh(d_2Q_{\|})}{\cosh[Q_{\|}(d_1+d_2)]-\cos[Q_{\perp}(d_1+d_2)]} . \tag{2}$$

Here d_1 and d_2 are the thicknesses of the magnetic and nonmagnetic layers, respectively, and $Q_{\|}$ and $Q_{\perp}$ are the components of the magnon wave vector parallel and perpendicular to the layers. Furthermore, $Q_{\perp}$ must satisfy the inequalities $0\le Q_{\perp}\le\pi/(d_1+d_2)$, while $Q_{\|}$ is determined by the experimental scattering geometry. [We again stress that Eq. (1) is only valid if $\vec{Q}_{\|}\cdot\vec{M}=0$; otherwise numerical solutions[7] are required.] The upper and lower limits of w given in Eq. (2) correspond to $Q_{\perp}=0$ and $\pi/(d_1+d_2)$. When the layers are thick compared to $Q_{\|}^{-1}$, or if d_1 and d_2 are very different, the range of values allowed for w is small and Eq. (1) predicts a narrow band of frequencies. For samples with thin, equal layers, w is allowed to range between very small values and 1, thus giving rise to a broad band of allowed modes.

In addition to the type-(a) modes described by Eq. (1), Refs. 6 and 7 predict that for $d_1 > d_2$ (magnetic layers thicker than the nonmagnetic ones), the surface modes in each layer combine to form a singular excitation which has the characteristics of a surface mode in the layered system. This mode appears at a frequency given by

$$\nu = \gamma(H + 2\pi M) , \quad (3)$$

which is the same as that of a surface magnon on a half-space of magnetization M. [It is also the same as Eq. (1) when $w = 1$.]

Before discussing the type-(b) modes we note that in a light scattering study consideration must be given not only to the allowed frequency bands described above, but also to the density of states within the band and the strength of the coupling of each magnon mode to the light. Numerical calculations of these effects are presented in Ref. 7 for a few particular choices of scattering geometry and sample characteristics. We note here some of the general trends. When $Q_{\|}d_1$ and $Q_{\|}d_2 \ll 1$, the density of states is largest for $Q_{\perp}(d_1+d_2) \sim \pi$. This indicates that the peak in the light scattering spectrum will appear at a frequency given by Eq. (1) with w near the minimum of its allowed range. As $Q_{\|}d_1$ and $Q_{\|}d_2$ increase, the density of states becomes more uniform over the allowed range of w, implying that the peak will appear close to the middle of the range of allowed w values and also that it is expected to be somewhat broader. For samples in which $Q_{\|}d_1$ and $Q_{\|}d_2 \gg 1$, the range of w is small and a well-defined peak is again expected.

The type-(b) modes arise from the interaction of standing spin waves in each layer. These modes are predicted[7] to lie at a frequency

$$\nu = \gamma[H(H + 4\pi M)]^{1/2} . \quad (4)$$

However, it is known from the study of standing spin waves in thin films[9,10] that the exchange interaction modifies this frequency considerably. In a single thin film of thickness d, the modes appear at frequencies given by

$$\nu = \gamma\{[H + D(n\pi/d)^2][H + 4\pi M + D(n\pi/d)^2]\}^{1/2} , \quad (5)$$

where D is the spin-wave stiffness constant and n is an integer. Hence, it is likely that these modes would not lead to a single peak as predicted by Eq. (4), but rather to a series of peaks at higher frequencies. The coupling of this type of mode to the light requires further theoretical investigation.

III. EXPERIMENT

The samples used in this study have been extensively characterized and described elsewhere.[11] They are composed of Ni and Mo layers of thickness d_1 and d_2, respectively, and consist of three series $d_1/d_2 = 3$, 1, and $\frac{1}{3}$. The range investigated is $180 \le d_1 + d_2 \le 10\,000$ Å. The saturation magnetization M_s at 10 kG and Curie temperatures determined from Arrot plots are shown in Figs. 1(a) and 1(b), respectively, and have been determined using superconducting quantum-interference device (SQUID) magnetometry. In this figure, triangles, crosses, and circles correspond to $d_1/d_2 = 3$, 1, and $\frac{1}{3}$, respectively.

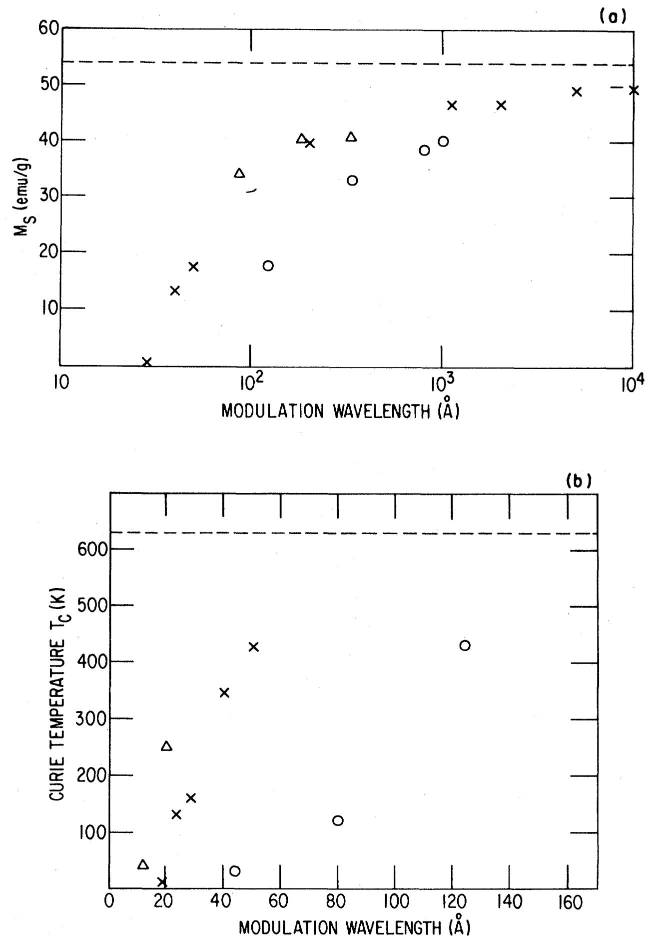

FIG. 1. (a) Magnetization and (b) Curie temperature for Mo/Ni superlattices. Triangles are for $d_1 = 3d_2$, crosses are for $d_1 = d_2$, and circles are for $3d_1 = d_2$. Dashed lines represent the values for bulk Ni.

The Brillouin spectra were recorded on a five-pass Fabry-Perot interferometer using $\sim$100 mW of 5145-Å radiation from an Ar laser. For a summary of previous experimental and theoretical work in the field of Brillouin scattering from magnons, the reader is referred to the review article by Borovik-Romanov and Kreines.[12] The scattering geometry is shown in Fig. 2. In our experiments, unless otherwise stated, the applied magnetic field is in the plane of the samples and perpendicular to the

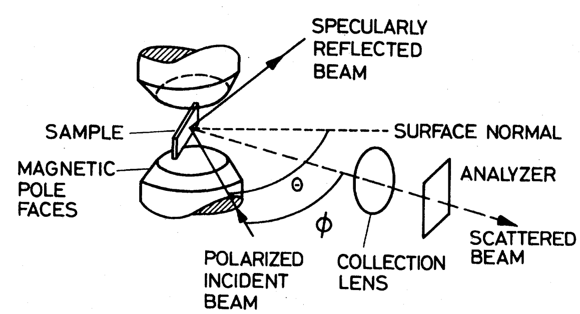

FIG. 2. Schematic diagram of the experimental arrangement used in our experiments.

horizontal scattering plane, the incident laser beam is horizontally polarized and makes an angle of 65° with the sample normal, the center of the collection optics makes an angle of 25° with the sample normal, and a vertical analyzer was placed in scattered beam. All the samples investigated were chosen so that the outermost layer would be nickel. The spectra were recorded with the samples at room temperature in fields between ~0.5 and 5 kG.

IV. RESULTS AND DISCUSSION

In Fig. 3 we present magnon spectra obtained from Mo/Ni superlattices. These spectra represent typical results for cases in which $d_1=3d_2$, $d_1=d_2$, and $3d_1=d_2$. As has been previously reported,[8] samples with $d_1=3d_2$ show two peaks at low fields ($\lesssim 2$ kG) which merge and become unresolved at higher fields. Samples in which $d_1=d_2$ usually show one peak; however, in a few instances to be discussed, a doublet is observed. Samples with $3d_1=d_2$ always show a single peak.

Figure 4 shows (crosses) the experimentally measured magnon frequencies versus applied field for a representative set of samples. The solid lines represent fits to the data using Eq. (1) for samples with $d_1 \le d_2$, and Eqs. (1) and (3) for samples with $d_1 > d_2$. M and w are used as fitting parameters, and the gyromagnetic ratio ($\gamma = 3.09$ GHz/kG) is taken from the literature.[13] According to the arguments given in the preceding section the data are analyzed in the following manner. For the case $d_1=3d_2$, where two peaks are observed, the higher-frequency mode is fit according to Eq. (3) by adjusting M. Then, using this value of M, the lower-frequency mode is fit with Eq. (1) using w as a parameter. Samples with $d_1=d_2$ and $3d_1=d_2$ which showed a single peak were fitted with Eq. (1) using M and w as parameters. Since both M and w could be varied, these fits led to larger errors in the estimates of M and w. The exception of this fitting procedure is the sample with $d_1=d_2=160$ Å. This sample showed a very broad peak or a doublet at the lowest fields, suggesting that the density of states was appreciable throughout the band described by Eq. (1). This is consistent with the

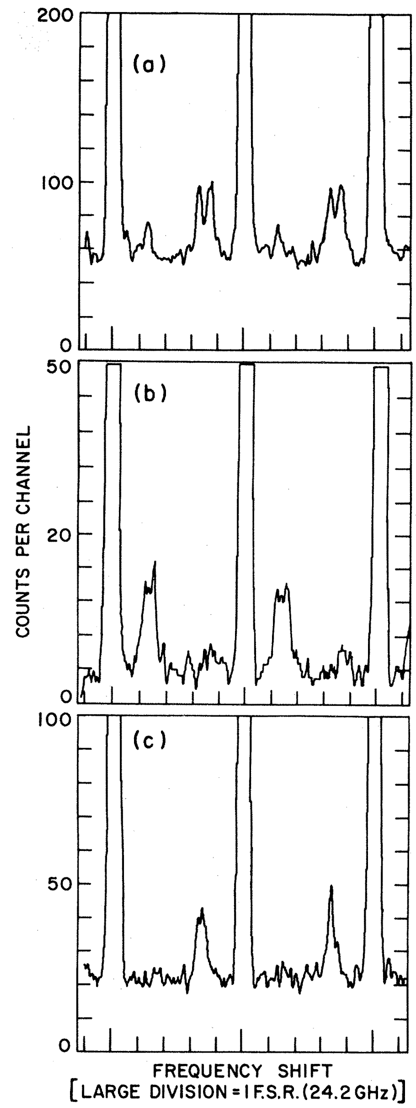

FIG. 3. Magnon spectra obtained from Mo/Ni superlattices with (a) $d_1=3d_2=249$ Å, applied field $H=+0.93$ kG; (b) $d_1=d_2=160$, Å, $H=-0.83$ kG; and (c) $3d_1=d_2=300$ Å, $H=+0.93$ kG. The direction of the frequency shift in (b) is different from that in (a) and (c) because the field direction was reversed.

TABLE I. Magnetization of Ni layers in a Mo/Ni superlattice obtained from dc-magnetization measurements and fits to magnon data. w values are from Eq. (2) and fits to the data (na denotes not applicable).

Sample d_2 (Å) Mo	d_1 (Å) Ni	w(fit)	w_{min}	w_{max}	M(fit) (kG)	M_s(dc magn.) (kG)
5000	5000	0.93 ±0.1	1.000	1.0	0.48±0.01	0.44±10%
1000	1000	0.60 ±0.03	0.747	1.0	0.44±0.03	0.42
550	550	0.76 ±0.03	0.381	1.0	0.45±0.02	0.42
160	160	na	0.043	1.0	0.38±0.02	0.38
100	100	0.045±0.07	0.017	1.0	0.39±0.02	0.36
750	250	0.30 ±0.10	0.256	0.775	0.45±0.03	0.35
600	200	0.80 ±0.05	0.177	0.766	0.33±0.02	0.34
300	100	0.18 ±0.06	0.049	0.754	0.36±0.02	0.29
180	540	0.15 ±0.05	0.152	0.764	0.35±0.02	0.41
83	249	0.07 ±0.03	0.036	0.753	0.30±0.02	0.36
46	138	0.03 ±0.03	0.011	0.751	0.28±0.02	0.36

calculations of Camley *et al.*,[7] which show that in some cases this band can exhibit broad structural features. In this case we fit the peaks with Eq. (1) using the maximum and minimum allowed values of w and using M as a fitting parameter.

Table I shows the values of M and w obtained from our fits, the values of M_s obtained from dc-magnetization measurements, and the range of allowed w values as determined from Eq. (2). For the fields used in our Brillouin measurements (0.5–5 kG) we expect the magnetization M to be equal to the saturation magnetization M_s to better than $\sim 1\%$. However, we should point out that the magnon positions depend on the length of time that the sample has been exposed to air. Samples exposed for over a month produced values of M as much as 30% lower than that from freshly prepared samples or samples that had recently been peeled from the substrate. The values presented in Table I were obtained from freshly peeled samples and were found to be reproducible to within $\sim 2\%$. Given the additional complication that magnon spectra from pure Ni are known to be very sensitive to surface preparation,[13,14] the values of the magnetization determined from dc magnetization and Brillouin scattering can be taken to be in agreement. The values obtained for w (especially those from samples with $d_1 = 3d_2$ for which the errors are small) are close to the low end of the allowed band, as predicted theoretically.[7] The one exception to this trend is the sample with $d_1 = 200$ Å and $d_2 = 600$ Å; we have no explanation for this behavior.

In the course of this work we have also studied the wave-vector dependence of the magnon frequencies. As expected, when two modes are observed, the upper one

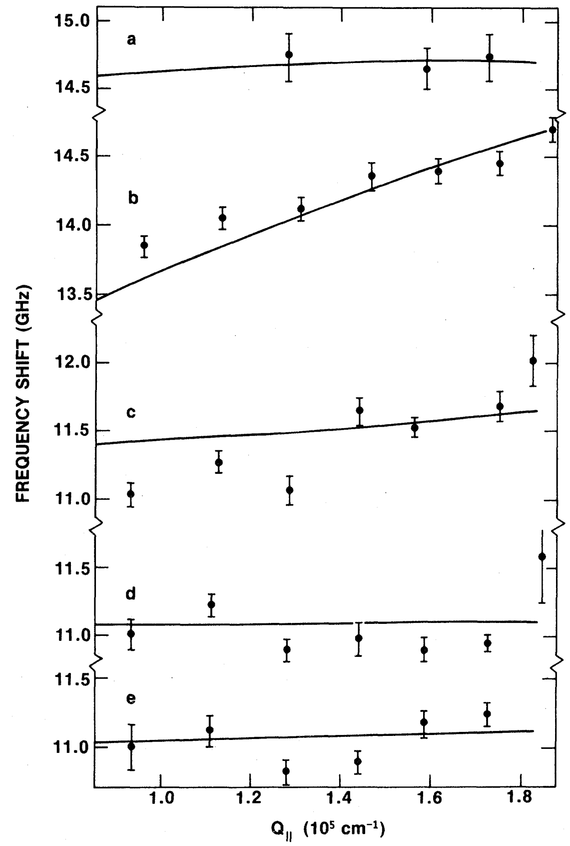

FIG. 5. Wave-vector dependence of magnons in a representative set of Mo/Ni superlattices. Solid lines are fits according to Eqs. (1) and (2). Magnetic field was 2.11 kG. The samples were as follows: *a*, $d_1 = d_2 = 2500$ Å; *b*, $d_1 = d_2 = 550$ Å; *c*, $d_1 = d_2 = 250$ Å; *d*, $d_1 = d_2 = 100$ Å; and *e*, $d_1 = 249$ Å, $d_2 = 83$ Å.

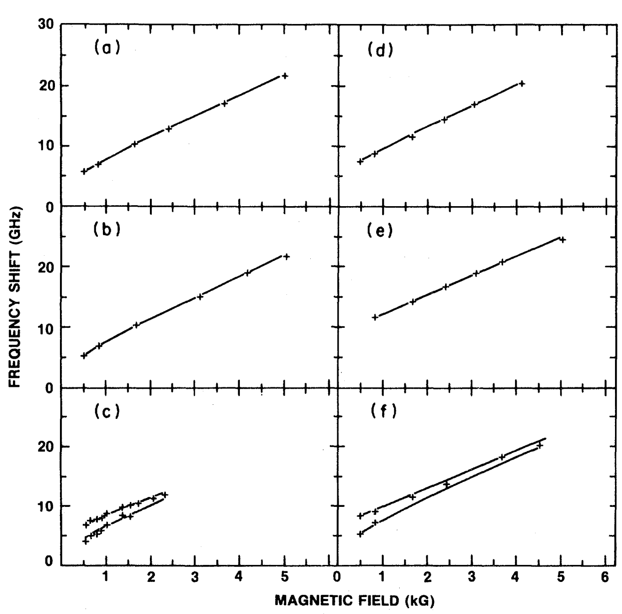

FIG. 4. Field dependence of magnon frequencies in a representative set of Mo/Ni superlattices. Solid lines are fits according to Eqs. (1) and (3). The samples are as follows: (a) $d_1 = 100$ Å, $d_2 = 300$ Å; (b) $d_1 = 100$ Å, $d_2 = 100$ Å; (c) $d_1 = 138$ Å, $d_2 = 46$ Å; (d) $d_1 = 250$ Å, $d_2 = 750$ Å; (e) $d_1 = 5000$ Å, $d_2 = 5000$ Å; and (f) $d_1 = 540$ Å, $d_2 = 180$ Å.

described by Eq. (3) is found to be independent of $Q_{\parallel}$. The position of the lower mode, or the single mode when only one peak is observed, is shown for a few samples in Fig. 5 as a function of $Q_{\parallel}$. The error bars are based only on the reproducibility of the data. The solid lines are fits according to Eqs. (1) and (2) with $Q_{\perp}(d_1 + d_2) = \pi$ and $H = 2.11$ kG. It can be seen from Fig. 5 that the experimental results are qualitatively explained by the theory. The discrepancy between theory and experiment observed in Fig. 5(c), can be explained as being due to our approximation $Q_{\perp}(d_1 + d_2) = \pi$. A full numerical analysis of the type performed in Ref. 7 would be necessary in order to draw quantitative conclusions.

As a final point, we mention that the sample $d_1 = d_2 = 550$ Å showed an additional mode at higher frequencies. The field dependence of this mode is shown in Fig. 6. Since this peak cannot be fitted with Eqs. (1) and (3) without resorting to unrealistic values of M and w, we conclude that the lower mode is of the type described by Eq. (1) (the values of M and w given in Table I refer to this mode), while the upper mode is of type (b), i.e., a combination of standing spin waves in each film. Fitting this mode according to Eq. (5), using M determined from the lower mode and $D(n\pi/L)^2$ as a fitting parameter, we obtain the upper solid line in Fig. 6. The fit yields $D(n\pi/L)^2 = 1.48$ kG, from which using the known value

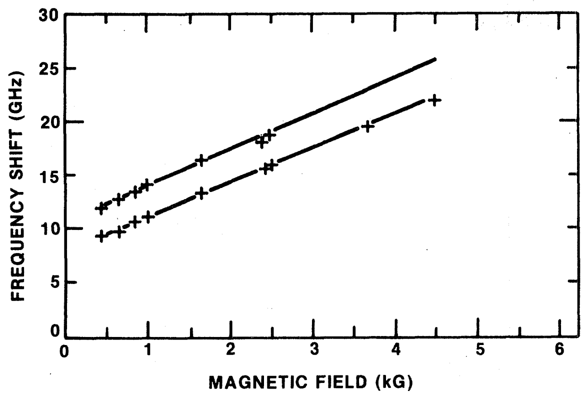

FIG. 6. Field dependence of the two modes observed in a superlattice with $d_1 = d_2 = 550$ Å. The lines are fits according to Eqs. (1) and (5).

of D for Ni [3.56×10^{-12} kG cm^2 (Ref. 15)], we obtain $L/n = (490\pm30)$ Å. Recalling that $d_1 = 550$ Å, this result indicates that we are apparently seeing the lowest standing spin wave mode in each layer and that coupling between the layers does not drastically modify its position relative to that in a single isolated film. Whether this is fortuitous or implies that Eq. (5) is a good approximation in all cases is not known. Unfortunately we were not able to follow as a function of field the additional peaks observed in other samples (notably the sample with $d_1 = 540$ Å and $d_2 = 180$ Å).

V. CONCLUSIONS

A study of magnetic excitations in a magnetic-nonmagnetic superlattice has been performed. Our results are found to be consistent with present theories, except, of course, in the case of modes which are known to be strongly dependent on exchange, which has not been included in the theory. Quantitative comparison between theory and experiment is not possible because of the numerical nature of the theoretical calculations, and also because of extraneous effects possibly arising from surface contamination.

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