Spin-wave modes in antiparallel magnetized ferromagnetic double layers

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For the first time the spin-wave-mode spectra of a symmetric dipolar coupled ferromagnetic double-layer system have been measured and compared in both the parallel and the antiparallel magnetized state by means of inelastic light scattering by use of a Brillouin-type spectrometer. Two symmetric Permalloy ($Ni_{80}Fe_{20}$) films of 25 nm thickness have been prepared with equivalent easy axes of an M-induced uniaxial anisotropy, but with different coercivities, $H_{C1}=9.2$ Oe and $H_{C2}=30.4$ Oe. By proper choice of remagnetizing fields applied along the easy axes, the parallel magnetized state (PM), $M_1+M_2=2M$, and antiparallel magnetized state (APM), $M_1-M_2=0$, could be realized. For both states the attributed spin-wave modes have been studied and compared with theoretical predictions. In particular, the predicted asymmetric, high-energy spin-wave modes of the APM state lying in the microwave frequency range (15 GHz) were shown to exist, to agree well with theory, and to be switched on and off reversibly by relatively small remagnetizing fields (15 Oe).

I. INTRODUCTION

Since their introduction by Bloch,[1] spin waves (magnons) have become increasingly interesting and have enabled a more profound understanding of ferromagnetism. In addition, since spin waves are propagating excitations, like electromagnetic waves and sound waves, they have been applied already in electronics as well as in communication techniques. The basic advantages of these magnetic modes are their tunability by an applied magnetic field and their slow propagation velocity (about five orders of magnitude smaller than that of electromagnetic waves).[2,3] Some devices, which make use of spin waves, such as delay lines, oscillators, and tunable filters, have already been produced commercially. New materials, like compositionally modulated films, which also exhibit spin-wave modes, are currently being developed and offer a considerable new potential for research and application.[4,5] Due to the proper choice of the component materials and of the numbers and thicknesses of layers, a tailoring of material properties and their basic magnetic excitations is now possible. A collective spin-wave-mode spectrum in a compositionally modulated multilayer system of parallel magnetized (PM) layers was predicted theoretically and proven experimentally soon afterwards by Brillouin scattering.[6-9] Recently, Mika and Grünberg[10] extended these calculations on PM multilayers to a multilayer system with alternating direction of film magnetization, i.e., to an antiparallel magnetized (APM) multilayer. They predicted new spin-wave modes and dispersion relations, which differ largely from those of the PM multilayers. In addition, an asymmetry for the Stokes and anti-Stokes magnon frequency distributions has been predicted. These results are of particular interest for both basic research and possible applications in microwave devices. However, so far no experimental evidence has been given because of the basic difficulties in realizing an equivalent symmetric ferromagnetic multilayer, which can be switched reversibly between the PM and APM states. Here we report on what maybe the first realization of such a ferromagnetic double layer, which can be properly switched between the PM and APM states and, hence, allowed the observation of the attributed spin-wave modes by Brillouin scattering. Since this magnetic double-layer structure (see Fig. 1) establishes the basic period for an APM multilayer structure, it is of particular interest for future investigations.

First we will outline theoretical results on our magnetic double layer for both the PM and APM states. Then we will report our experiments and results on the spin-wave modes for the two different magnetization states.

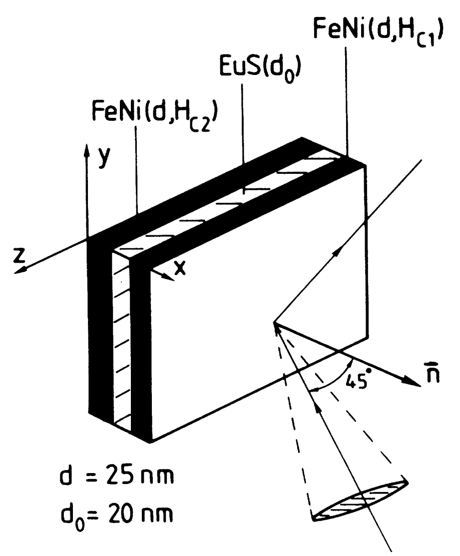

FIG. 1. Structure of the ferromagnetic double layer and the light scattering geometry.

II. DIPOLAR SPIN WAVE MODES OF A FERROMAGNETIC DOUBLE LAYER

Following the Damon and Eshbach (DE) calculation[11] for a single ferromagnetic layer, Grünberg[5] extended the theory to a ferromagnetic double- and multilayer system in the PM and APM states. For later comparison with our experiments only the results on the dispersion relation of a symmetric double layer consisting of two equivalent, equally thick ferromagnetic films are considered and presented here, i.e.,

$$2-e^{-2kd}-e^{2kd}-e^{2kd_0}\left[2+Ae^{-2kd}+\frac{1}{A}e^{2kd}\right]=0\ . \qquad (1)$$

Here d and d_0 are the thicknesses of the magnetic and nonmagnetic layers, respectively, k is the wave vector of the spin wave, and A is defined as

$$A=(\kappa^2-\nu^2)/(\nu-\kappa-2)(\nu+\kappa+2) \qquad (2)$$

and

$$\kappa=\frac{\Omega_H}{\Omega_H^2-\Omega^2}\ ,\quad \nu=\frac{\Omega}{\Omega_H^2-\Omega^2}\ ,$$
$$\Omega_H=B_0/J_0=H/M\ ,\quad \Omega=\frac{\omega}{\gamma J_0}\ . \qquad (3)$$

In Eqs. (2) and (3), B_0 is the applied magnetic field, J_0 the static magnetization, γ the gyromagnetic ratio, and ω the frequency of the corresponding spin waves.

Equation (1) has two solutions, which are shown schematically in Fig. 2 by the solid lines. It is obvious that for large values of kd_0, the dipolar coupling is very weak. Then one obtains the doubly degenerated DE mode for the two decoupled layers. However, with reduction of the interlayer thickness d_0 or by increasing the wavelength of the spin wave, the dipolar coupling becomes stronger. The initially degenerate DE modes are now split into two modes, one with higher, the other with lower, frequency. As kd_0 approaches zero, the upper branch becomes the surface mode of the film with thickness $2d$, while the lower branch merges with the volume mode of the $2d$ film. For both the Stokes and the anti-Stokes lines, the behavior is the same.

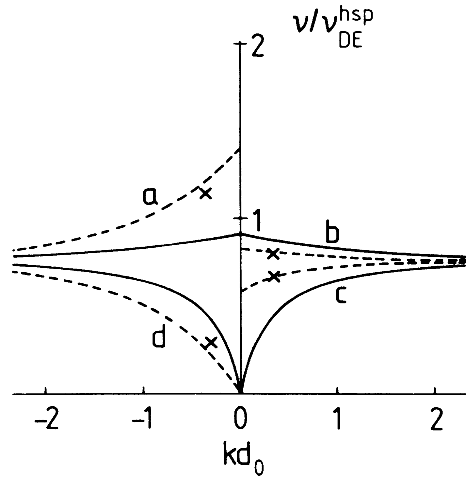

FIG. 2. Theoretical spin-wave dispersion branches of the ferromagnetic double layer with $d_1=d_2=25$ nm, $k_yd_{1,2}=0.43$, $d_0=20$ nm, $k_yd_0=0.34$, and $J_{1,2}=1$ T, shown in Fig. 1 for the parallel magnetized (PM, $J_1=J_2$, solid curve) and antiparallel magnetized (APM, $J_1=-J_2$, dotted curve) state. The experimental results (×) are shown for the APM state 2 of Fig. 4. (Note that all three states, 1, 2, and 3 in Fig. 4 are retained after switching off the applied field, which allows us to compare directly with theory.)

For the APM situation and with zero applied magnetic field, Mika and Grünberg[10] deduced the dispersion relation for an N-layer system. Two interesting points only should be emphasized here. Firstly, due to the antiparallel coupling between the adjacent layers, a bulk mode band emerges, in which the highest-frequency value is about double that of the normal single-layer DE modes. For zero applied magnetic field it would be interesting if one could reach such a high frequency for the spin waves. Secondly, these spin-wave modes show an asymmetric frequency distribution for Stokes and anti-Stokes lines, if the layer number N is even (see Fig. 3 in Ref. 10). For our case of $N=2$, the dispersion relation can be deduced from the following determinant:

$$\begin{vmatrix} 1 & (1+2\Omega)e^{kd} & 0 & 0 \\ (1-2)e^{kd} & 1 & (1+2\Omega)e^{-kd_0} & e^{k(d-d_0)} \\ e^{k(d-d_0)} & (1+2\Omega)e^{-kd_0} & 1 & (1-2\Omega)e^{kd} \\ 0 & 0 & (1+2\Omega)e^{kd} & 1 \end{vmatrix}=0\ . \qquad (4)$$

Here, all parameters have the same meaning as defined before. Equation (4) has four solutions corresponding to four branches of spin-wave modes. For comparison with the PM state (solid lines of Fig. 2), the experimental results for the APM state and their dispersion relations are shown in Fig. 2 by the crosses and the dotted lines, respectively.

When kd_0 is large, these modes are the degenerate normal single-layer DE modes such as in the case of the PM state. With decreasing interlayer thickness more coupling is introduced and causes the DE mode splitting. Obviously, the splitting in the APM state is different from that in the PM state. For one of the propagation directions related to the dotted curves of Fig. 2, the splitting takes place earlier and is larger for the same kd_0, as compared to the other direction and with the PM states (solid curves of

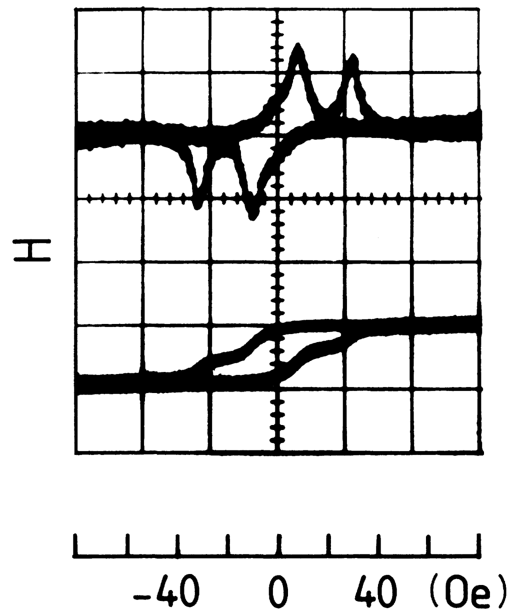

FIG. 3. 50-cps hysteresis loop (lower part, reproduced enlarged in the upper part of Fig. 4) of the $Ni_{80}Fe_{20}$ double layer of Fig. 1 and its derivative (upper part). They indicate the different coercive forces, H_{C1} and H_{C2}, needed to achieve the intermediate APM state 2 between the PM states 1 and 3 of Fig. 4.

Fig. 2). The splitting of the other propagating direction is even smaller than that of the PM state. The highest frequency of these modes reaches a value of approximately double that of the normal DE mode of an infinitely thick sample, i.e., the surface spin-wave mode of a ferromagnetic half space ($\nu_{\mathrm{DE}}^{\mathrm{hsp}}$ in Fig. 2).

III. EXPERIMENTAL PROCEDURE

A. Film preparation

Two material properties can be used to achieve the antiparallel alignment of the magnetization of the two magnetic layers. One of these makes use of the difference in the coercive forces between the two layers. The other one is based on a possible antiferromagnetic coupling between the two ferromagnetic layers through an interlayer.[10] The latter method has been realized in our laboratory recently, also.[12]

It is well known that the coercive field H_C of a film is very sensitive to composition, structure, doping, defects, and even to the substrate and coverage layers. However, this complication also offers possibilities for achieving different coercive forces in a double- or multilayer system. In our case, for two ferromagnetic Permalloy layers ($Ni_{80}Fe_{20}$) of equal thickness the coercive forces (H_{C1} and H_{C2}) could be made different, and with hysteresis loops of nearly rectangular shape for remagnetization along their uniform magnetic easy axes. Then, with the applied field kept aligned in the M-induced easy axes of the Ni-Fe films, the APM situation could be realized in the following way: Firstly, the films were saturated in one direction by applying a sufficiently large magnetic field. Secondly, the field was reduced to zero and then reversed in sign. When increasing the reversed field amplitude until the smaller value H_{C1} was achieved, the magnetic weak layer 1 became reversed and the APM state (2) was realized. It was retained even after switching off the applied field (see Fig. 4).

For quantitative comparison with theory, we have

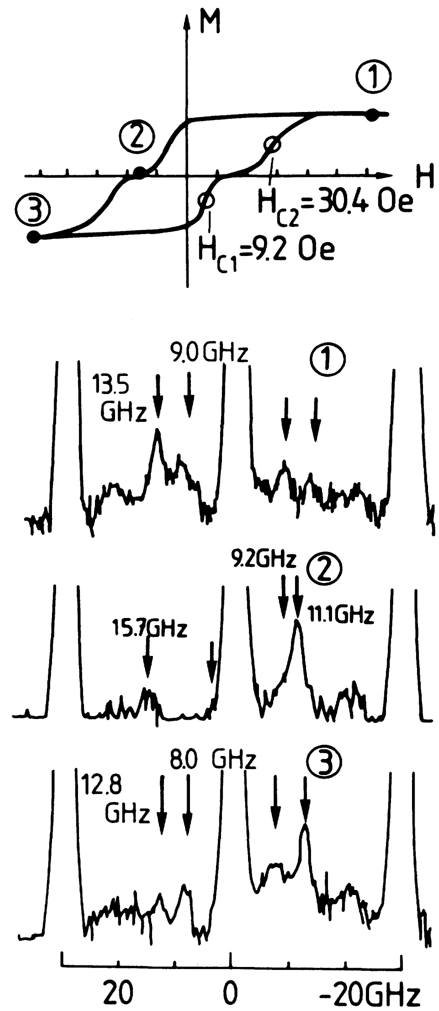

FIG. 4. Spin-wave spectra measured at room temperature by Brillouin light scattering on the magnetic double layer of Fig. 1 in the PM states 1 and 3, and in the APM state 2. The arrows indicate the positions of the DE spin-wave lines observed for $B_0 = +0.0185$ and -0.0185 T with the PM states 1 and 3, respectively, and for $B_0 = -0.002$ T with the APM state 2. At these B_0 values they are shown as measured results (crosses) in Fig. 5 to be compared there (and also in Fig. 2) with theoretical B_0 and d_0 dependences deduced from Eqs. (1)–(3) for the PM states 1 and 3 and from Eqs. (2)–(4) for the APM state 2.

TABLE I. Samples *a* and *b*, compositions.

Substrate	Layer I	Interlayer	Layer II	Coverage
Sample *a*				
Material: Al_2O_3	$Fe_{20}Ni_{80}$	EuS	$Fe_{20}Ni_{80}$	
Thickness: 0.5 mm	250 Å	200 Å	250 Å	
Sample *b*				
Material: Al_2O_3	Fe	EuS	Fe	ZnS
Thickness: 0.5 mm	260 Å	300 Å	260 Å	500 Å

chosen a ferromagnetic double layer having the same magnetization and thickness for the two Permalloy layers, but sufficiently different coercive forces. This was achieved by using three different substrate temperatures, i.e., $T_s = 400$ and 200 °C for the Permalloy layers and $T_s = 300$ °C for the EuS intermediate layer. In Table I we have listed some data of the Ni-Fe film systems prepared successfully in this way. Also included are data for an equivalent Fe double-layer system, which has been studied for comparison.

Epipolished sapphire (Al_2O_3) was used as the substrate. EuS, which is paramagnetic at room temperature (Curie temperature 16 K) and has a cubic structure, was used as the insulating interlayer. Sometimes a ZnS layer was deposited on top of the film to improve the optical surface properties and to protect the metal film against oxidation, which, however, is not critical in the case of Permalloy films.

All the films were prepared at ultrahigh-vacuum conditions. During evaporation the residual pressure increased to about 5×10^{-8} Torr. Electron-beam heating was used for evaporating Permalloy, Fe, and EuS, while ZnS was deposited from a resistance-heated crucible. The evaporation rate was controlled by a vibrating quartz system and kept typically at 0.1 nm/s for Permalloy and Fe.

B. Magnetization studies

Two methods, i.e., an inductive method[13] and the method based on the magneto-optic Kerr effect[14] (MOKE) have been used to characterize the magnetization state of the double layer and at the same time to measure the coercive forces. Figure 3 shows a typical result obtained by the inductive method for sample *a* of Table I. Obviously the hysteresis loop is composed of two square loops. The two different magnetization reversals and attributed H_{C1} and H_{C2} values are indicated by the different peaks of the differentiated hysteresis signal shown in the upper part of Fig. 3. From these measurements the two indicated coercive forces were deduced. However, one could argue that the two peaks at $H_{C1} = 9$ Oe and $H_{C2} = 34$ Oe could also originate from two phases of a single layer. In order to prove that the two peaks are really due to the magnetization reversal of the two different Permalloy layers, the MOKE has been used. It is an ideal method for probing the distinct coercive forces of the single films since the penetration depth of visible light in Fe or Permalloy is only about 10–20 nm, which is just the thickness of one layer. In our case the two magnetic layers are all accessible by light, since both the substrate (Al_2O_3) and the coverage (ZnS) are transparent to visible light. The MOKE measurements showed, in fact, that the first-deposited NiFe layer, i.e., that between Al_2O_3 and EuS and grown at $T_s = 400$ °C, had the larger H_C value, H_{C2}, while the top Permalloy layer, i.e., that between EuS and ZnS and grown at $T_s = 200$ °C, had the lower H_C value, H_{C1}, and, hence, its magnetization was reversed first.

C. Light scattering experiments

A tandem multipass Fabry-Perot interferometer was used for the Brillouin scattering experiments. The details of the Brillouin spectrometer and light scattering from spin waves have been described elsewhere.[15,16] We report only briefly the experimental conditions. An Ar^+ laser of 514.5 nm wavelength was used as the light source. The laser power incident on the sample surface was about 50–100 mW.

In a backscattering geometry (see Fig. 1), the incident beam formed an angle of 45° with the normal of the sample surface. The Ni-Fe films were magnetized along the in-plane easy axis. The sample magnetization directions thus were always perpendicular to the light beam and parallel to each other. In this arrangement the detected magnons have a k vector of 1.72×10^5 cm^{-1}. We measured the spin-wave modes by Brillouin scattering for three different magnetization states labeled (1), (2), and (3) in Fig. 4. First, by applying a high magnetic field, the PM state (1) of the double film is obtained. In this state the magnetic field dependence of the magnon frequency as shown in Fig. 5 was measured. Then we reduced the applied magnetic field to zero, and increased it with opposite sign up to -20 Oe (in the case of sample *a* in Table I). This small opposite field reversed the magnetization of layer II and, thus, produced the APM state (2) of the double layer. In this state we then measured again the spin-wave modes as indicated by spectrum (2) in Fig. 4. When a large opposite magnetic field was applied subsequently, the magnetization of both films became saturated in the opposite direction, i.e., state (3) as compared to the initial state (1). The measured spin-wave modes of spectrum (3) in Fig. 4 then have the same energies, while the line intensities of the Stokes and anti-Stokes lines are reversed as compared those of spectrum (1).

IV. RESULTS AND ANALYSIS

Figure 4 shows typical spectra measured by Brillouin scattering on sample *a* of Table I. The results correspond

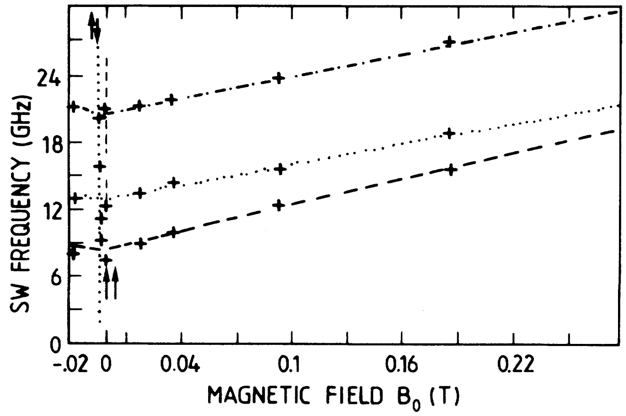

FIG. 5. Magnetic field dependence of the magnon frequencies measured from the magnetic double layer of Fig. 1 in the PM state. It fits well to the theory by Grünberg and Mika (Ref. 10) with the material parameters given in Table II and Fig. 2. The vertical dotted line at small negative fields indicates the line shifts due to the transition of the double layer into the APM state.

to the PM states (1) and (3), and the APM state (2), as indicated.

It should be pointed out that all the light scattering experiments were performed with crossed polarizer and analyzer. Thus, the phonon peaks could be suppressed and the recorded peaks were due to magnons only, as proved also by the magnetic field dependence of the magnon frequencies.

By measuring a series of spectra at different applied magnetic fields for the PM state, the results shown in Fig. 5 were obtained. Measured points have been fitted by the theoretical model for double layers and standing-spin-wave (SSW) modes. Three kinds of peaks were recorded. One was due to the lowest-energy SSW mode ($n=1$) of the double film, which follows the dispersion relation:

$$\omega_{SSW}=\gamma\left\{\left[D_{ex}\left[n^2\frac{\pi^2}{d^2}+k_y^2\right]+\frac{J}{2}+B_0\right]^2-\left[\frac{J}{2}\right]^2\right\}^{1/2}, \quad (5)$$

where γ is the gyromagnetic ratio, D_{ex} is the exchange constant, n is the number of modes of the SSW indicating the sequence of increasing energy, and k_y is the wave-vector component perpendicular to the direction of the magnetic polarization vector **J** and to the magnetic field vector B_0. Taking the dipolar interaction into account, the standing spin-wave line of a magnetic double layer will also be split. However, according to Vaihinger and Kronmüller[17] this splitting is normally very small (for most cases, less than 0.5 GHz, which is difficult to detect in Brillouin scattering). So, for simplicity, the dispersion relation of SSW modes from magnetic single layers is presented here.

The other two peaks are due to the splitting of the DE modes of the magnetic double layer in the PM state. They follow the dispersion relation predicted by Eq. (2). These assignments are confirmed by the good agreement found between the experimental theoretical spin-wave energy data. For both SSW and split DE modes, the same parameters (J,D_{ex},γ) were used in fitting the measured points in Fig. 5. On the other hand, the comparison between our fitting parameters and the results taken from the literature (see Table II) shows that both our light scattering measurements and the quality of our magnetic double layer are reliable. On this basis we performed the light scattering experiment with the sample in the APM state (2). The only variation made to achieve this state was the proper setting of the reserved magnetic field.

As can be seen from spectrum (2) corresponding to the APM state in Fig. 4 this variation results in a qualitative change as compared to the spin-wave spectrum (1). Comparing spectrum (2) for the APM state and spectrum (1) or (3) recorded for the PM state, it become obvious that the peak positions of spectrum (2) are no longer symmetric on the Stokes and anti-Stokes side. Instead, in the APM spectrum (2) one of the two split DE modes on the one side seems to disappear, while simultaneously a new mode at a higher frequency (15.7 GHz) emerges. On the other side, now only one pronounced and sharp peak instead of the two is observed. This sharp peak shows an asymmetric line shape, indicating that, in fact, it is composed of two unresolved peaks. This means, that their splitting has become smaller than in the PM state (1).

In Table III we have listed for comparison the measured mode frequencies for the APM state and those expected by using the theory of Mika and Grünberg.[10] The latter assumes $B_0=0$, which, however, is in agreement with our experimental situation since the fields $H=H_{C1,2}$ are negligibly small or can even be switched off without changing the magnetization states (1), (2), and (3). Although with the Permalloy double layer one spin-wave mode could not be observed because its energy was too small, the three measured modes are in good agreement with the prediction of theory.

It should be pointed out that the sharp peaks in spectrum (2) are due to two surface modes propagating in the same direction, and with the indicated smaller frequency difference, thus, displaying the stronger intensities. Along the other direction two volume modes are propagating. Their scattering intensities are found to be much weaker; however, their frequency difference is much larger than that of the two surface modes. For large k_yd and $k_yd_0\to 0$ the highest value of $\nu/\nu_{DE}^{hsp}=2.0$ is predicted from theory. For our experiment with $k_yd=0.43$ and $k_yd_0=0.34$ the predicted value was 1.22 (see Table III and Fig. 2), while the measured value was 1.14. Normally, the DE surface mode is called "nonreciprocal," because it propagates only in one direction, which is deter-

TABLE II. Parameters obtained by Brillouin scattering measurements in the PM state of sample a, and data from Ref. 18 for comparison.

	J (T)	A (10^{-11} W s/m)	g
Our results	1.00	0.96	1.98
Literature[a]	1.06	0.80	2.01

[a]Reference 18.

TABLE III. Comparison of experimental and theoretical results.

	ν/ν_{DE}^{hsp} [b]	Normalized SW frequencies for the APM state[a] a	b	c	d
$Ni_{80}Fe_{20}$ (Permalloy)					
	Theory	1.22	0.81	0.66	0.25
	Experiment	1.14	0.80	0.67	
Fe					
	Theory	1.16	0.81	0.70	0.34
	Experiment	1.01	0.82	0.70	0.28

[a]See Fig. 2.
[b]ν_{DE}^{hsp} is the frequency of the DE mode of a half-space.

mined by the normal of the surface and the direction of the applied magnetic field. This property can be used in nonreciprocal devices. However, at the other surface of the film a wave can propagate with the same frequency but in the opposite direction. This obscures the nonreciprocal property of the DE mode in our case. However, for a double layer in the APM state (2) this problem becomes ruled out completely because now the two surface waves propagate only in one direction, while in the other direction only volume spin waves with different frequencies are propagating. These differences in the PM- and APM-state spin-wave modes and in their propagation behavior may be of interest in device applications.

No such drastic change of the spin-wave energy has been observed for the SSW modes. However, we found that the scattering intensities are different on the Stokes and anti-Stokes sides. In fact, the small intensity occurs on the sides where the volume modes propagate. This reduction of scattering intensity may be due to interference between these bulk modes.

Several other samples of composition (Fe/EuS/Fe) and different thicknesses of d(Fe) and d_0(EuS) have been prepared and studied. Only the Fe sample b of Table I exhibited behavior similar to the Permalloy sample a. The results are included in Table III. While for sample b the coercivities of the two Fe layers were different too, the hysteresis loops were not of rectangular shape as were those of the Permalloy sample a (see Fig. 3). The cubic instead of uniaxial in-plane anisotropies of Fe films, together with inhomogeneous interface stresses, may cause a less homogeneous antiparallel magnetization alignment in the APM state of the Fe films as compared to that of the Permalloy films. The preparation of more homogeneous single-crystal Fe films may improve results in the future (see, e.g., Ref. 12).

V. CONCLUSION

It has been demonstrated by means of a properly prepared $Ni_{80}Fe_{20}$ ferromagnetic double-layer sample that two magnetization states can be realized: the PM state with the magnetization of both layers aligned parallel, and the APM state with its magnetization aligned antiparallel.

For what we believe to be the first time, the characteristic differences expected from theory for the spin-wave modes of such a dipolar coupled double-layer system in these two magnetization states could be demonstrated experimentally by means of Brillouin spectrometry. The spin-wave mode of double energy in the APM state can be achieved from the initial PM state by applying only a small reversed field. Together with the change in symmetry and propagation behavior of the related spin-wave modes, this may be useful for microwave devices and other applications. An extension to ferromagnetic multilayer systems, with equivalent PM and APM magnetization states and the theoretically predicted[10] bandlike spin-wave mode spectra, seems a worthwhile and now realistic goal for further experimental efforts.

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