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Collective spin waves in Fe-Pd and Fe-W multilayer structures

B. Hillebrands, P. Baumgart, R. Mock, and G. Güntherodt
II. Physikalisches Institut, Universität Köln, 5000 Köln 41, Federal Republic of Germany

A Boufelfel and C. M. Falco
Department of Physics and Optical Sciences Center, University of Arizona, Tucson, Arizona 85721
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The complete, theoretically predicted band of collective magnetostatic spin-wave excitations has been observed for the first time in Fe-Pd and Fe-W multilayer structures by Brillouin scattering. Despite the large number of bilayers (49–90) discrete spin-wave modes due to a quantization of the momentum perpendicular to the stack have been observed in the upper half of the band. For Fe-Pd multilayer structures the saturation magnetization shows a monotonic decrease of up to 30% from that of the bulk Fe for modulation wavelengths down to 33 Å.

Metallic multilayered systems have attracted strong interest recently due to the novel possibility of engineering their physical properties over a wide range. However, most of the new properties observed to date have been related to thin single film effects or to the large number of interfaces. The only collective excitations which have been established so far are the theoretically predicted[1-3] collective magnetostatic spin-wave excitations in a stack of alternating magnetic and nonmagnetic layers. A band of a new type of collective spin-wave excitations is formed as a result of the mainly dipolar coupling between the surface [Damon-Eshbach (DE)] spin waves of each magnetic layer. Preliminary experimental evidence for these modes was obtained by Brillouin scattering in Mo-Ni multilayer structures.[4,5] However, an experimental proof of the predicted shape and detailed features of the excitation band has until now been missing.

In this paper we report on Brillouin scattering experiments in Fe-Pd and Fe-W multilayer structures with 49 to 90 bilayers. We observe for the first time the theoretically predicted shape of the collective spin-wave band, its partly bulk- as well as surface-mode-like character and well-resolved, discrete spin-wave modes in the upper part of the excitation band. The latter modes are observed due to the finite though large number of magnetic layers in the samples. For the Fe-Pd samples, the values of the saturation magnetization $4\pi M_s$ obtained from our measurements show a monotonic softening from 19 to 14 kG as the modulation wavelength L (bilayer thickness) is decreased from 188 to 33 Å. This is in contrast to the $L=81$ Å Fe-W sample, which within experimental error has the bulk value for $4\pi M_s$ of 21 kG.

The Fe-Pd and Fe-W multilayer samples were prepared on single-crystal sapphire substrates using a sputtering technique described elsewhere.[6] The sample parameters are listed in Table I. The samples were prepared with equal numbers of atomic layers N_{at} for the magnetic and nonmagnetic layers rather than with equal thicknesses per layer. The superlattice or modulation wavelengths of the different samples (Table I) were determined by x-ray diffraction. As shown by Bragg and wide-film Debye-Scherrer x-ray diffraction, the layers grew with a preferred orientation of bcc Fe(110) planes, fcc Pd(111) planes, and bcc W(110) planes, with no correlation of the in-plane orientation among subsequent layers. Long-range structural coherence of at least 300 Å perpendicular to the layers was exhibited by the samples.

The Brillouin scattering experiments were performed at room temperature under vacuum in backscattering geometry using a (3+3)-pass tandem Fabry-Perot interferometer.[7] A single-moded 5145-Å Ar^+-ion laser was used with an incident power of p-polarized light of up to 250 mW. The sampling time per spectrum was typically 5 h. The inelastically scattered light was analyzed depolarized for spin-wave analysis. The applied magnetic field was oriented parallel to the layer planes and perpendicular to the scattering plane.

Figure 1 shows a series of Brillouin spectra of the Fe-Pd multilayer structures (A–H) and of a Fe-W multilayer structure in an applied magnetic field of $H=1$ kG. The band of collective spin-wave excitations can clearly be identified by the strong asymmetry of the scattering intensity in the right-hand part of the spectrum. The density of states is largest at the lower band edge (smallest frequency shift) and decreases asymmetrically toward the upper band edge, where some additional discrete small peaks are

TABLE I. Parameters of the investigated Fe-Pd and Fe-W multilayer samples. N_{at} is the (equal) number of atomic layers in each magnetic and nonmagnetic layer.

Sample	Modulation wavelength L (Å)	Total bilayers	N_{at}	Total thickness (Å)
Fe-Pd A	33.2	90	8	2988
Fe-Pd B	40.1	90	9	3609
Fe-Pd C	46.2	90	11	4158
Fe-Pd D	64.6	70	15	4522
Fe-Pd E	77.8	70	18	5446
Fe-Pd F	86.8	70	20	6076
Fe-Pd G	89.2	51	21	4549
Fe-Pd H	188.4	49	44	9232
Fe-W	80.7	60	19	4840

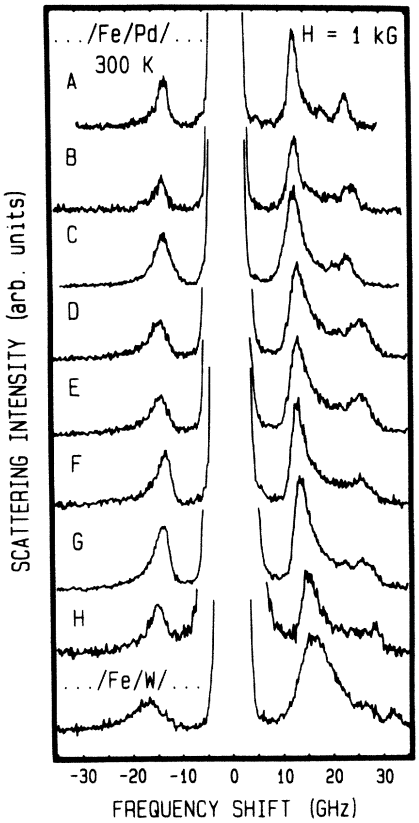

FIG. 1. Measured room temperature Brillouin spectra of Fe-Pd and Fe-W multilayer structures in an applied magnetic field of 1 kG. For Fe-Pd multilayer structures the modulation wavelength increases from A to H. For sample parameters, see Table I. The angle of incidence was 43°.

superimposed. If we neglect for a moment these small discrete peaks near the upper band edge, the shape of the spin-wave excitation band is qualitatively similar to the calculated Brillouin scattering cross section for the semi-infinite multilayer system Mo-Ni.[1] The asymmetry of the Stokes–anti-Stokes scattering intensities, especially for the small peaks near the upper band edge, is indicative of excitations with dominant surface-mode character.

Before discussing the additional discrete peaks observed in the spectra we briefly review the calculation of the mode frequencies of a semi-infinite multilayer structure.[1-3] In the magnetostatic limit for an infinite number of layers the frequencies ω of the band of excitations are given by

$$\omega=\gamma[H(H+4\pi M_s)+(2\pi M_s)^2 w]^{1/2} , \tag{1}$$

where γ is the gyromagnetic ratio, H the applied magnetic field, and $4\pi M_s$ the saturation magnetization. Exchange contributions and surface anisotropy are neglected.[1] The quantity w is defined by

$$w=\frac{2\sinh(q_\parallel d)\sinh(q_\parallel d_0)}{\cosh(q_\parallel L)-\cos(q_\perp L)} , \tag{2}$$

where $q_\parallel$ ($q_\perp$) is the spin-wave momentum parallel (perpendicular) to the layers, d (d_0) is the thickness of each magnetic (nonmagnetic) layer, and $L=d+d_0$ is the modulation wavelength. Using Bloch's theorem $q_\perp$ takes values in the interval

$$0\le q_\perp \le \pi/L . \tag{3}$$

This implies that for $d\approx d_0$ the upper band edge is at $w=1$, which coincides formally with the *DE* mode of a half-space. The lower band edge is at $w=\tanh^2(\frac{1}{2}q_\parallel L)$, yielding $w\approx 0$ for the parameters of Table I.

$q_\perp$ increases in going from the upper to the lower band edge and therefore the surface-mode-like character changes continuously to a bulk-mode-like character. This can be seen directly in Fig. 1 by the Stokes–anti-Stokes asymmetry, which is strongest at the upper band edge. This asymmetry was not observed in the Brillouin spectra of Co-Nb multilayer structures with 11 or less bilayers,[8] except for the highest frequency mode.

If the number of magnetic layers N is finite, it is obvious that $q_\perp$ is quantized into N discrete values $q_\perp^{(i)}$, $i=0,\dots,N-1$. However, any influence of this quantization on the spin-wave spectrum can only be observed if the frequency separation between the modes $q_\perp^{(i)}$ and $q_\perp^{(i+1)}$ is larger than their linewidths. This is surely the case for small numbers of magnetic layers as in Ref. 8 ($N\le 11$), where all discrete modes are observable. Nevertheless, it should also be possible to observe these quantized modes in samples with very large N if the modulation wavelength is sufficiently small as well as completely uniform throughout the sample. We can easily derive an estimate which demonstrates this. For $q_\parallel L \ll 1$, and $q_\perp L \ll 1$, Eq. (2) can be approximated by

$$w=\left[1+\left(\frac{q_\perp}{q_\parallel}\right)^2\right]^{-1} , \tag{2'}$$

where $q_\parallel$ is given in backscattering geometry by

$$q_\parallel=\frac{4\pi}{\lambda}\sin\theta , \tag{4}$$

with λ the wavelength of the light and θ the angle of incidence. A rough estimate of the quantization of $q_\perp^{(i)}$ is

$$q_\perp^{(i)}=\frac{i\pi}{NL},\ i=0,\dots,N-1 . \tag{5}$$

Thus Eq. (2') can be rewritten as

$$w_i=\left[1+\left(\frac{i\lambda}{4NL\sin\theta}\right)^2\right]^{-1} . \tag{2''}$$

To observe the $i=1$ mode and the $i=2$ mode with a frequency separation of, e.g., 20% at $\theta=45°$ requires $\lambda/NL\ge 1$. This implies large values for N are needed if the modulation wavelength L is small compared to λ. Hence, the modulation wavelengths used for the experiments reported here (33 Å $\le L \le$ 188 Å) require N smaller than 160 ($L=33$ Å) and 30 ($L=188$ Å). As can be seen from Table I this is the same order of magnitude as those used in our experiments. For sample H ($N=49$) no splitting should be observed between the $i=1$ and $i=2$ peaks. This is in agreement with Fig. 1, where only the $i=1$ peak is observable, whereas the $i>1$ peaks form an unresolvable continuum of modes. Thus, despite the large but finite number of bilayers the observed discrete peaks in most of our spectra can be attributed to well-separated spin-wave modes of the mutilayer structures. Out of the actually 90 discrete frequency values expected from the above discussion for samples A–C, the highest few (up to four, see below) can be experimentally resolved. The others form a continuum with a maximum of the density of states at the lower band edge.

To calculate the exact quantization of $q_\perp$ one has to evaluate numerically a recursion formula for the mode frequencies as derived by Grünberg and Mika.[2] If this is done one obtains qualitatively the same behavior but with slightly different values of $q_\perp^{(i)}$ compared to Eq. (5), due to the inclusion of the proper boundary conditions at the bottom and top layers of the stack.

In Fig. 2 the calculated mode frequencies (solid lines) for sample C are plotted as a function of the applied magnetic field. Also shown are the experimental values of the lower band edge (◇) and the discrete modes (△). As fitting parameters we used $4\pi M_s$ and γ, which are given by the intersection of the highest mode with the ordinate axis and by the slope of the uppermost curve, respectively. To achieve a slightly better fit we have added to the applied magnetic field an exchange field of 67 G, which corresponds to the bulk exchange constant of $D_{ex} = 2.45 \times 10^4$ Å^{2}kG.[9] The results of this fitting procedure are $4\pi M_s = 13.9$ kG and $\gamma = 19.0$ GHz/kG, where the latter corresponds to a g factor of 2.15. Within experimental error the measured values can be seen to agree well with the calculated curves over the entire range of applied magnetic field. The values obtained for $4\pi M_s$ and γ are also confirmed by additional measurements of the $q_\parallel$ dependence of the discrete modes.

In Fig. 3, the fitted values of $4\pi M_s$ are plotted as a function of the thickness d of the Fe layers. For the Fe-Pd multilayer structures the saturation magnetization decreases from 19 to 14 kG with d decreasing from 89 to 16 Å, whereas the Fe-W multilayer structure shows a value of $4\pi M_s = 20.1$ kG which agrees within the error bars with the bulk value for Fe. As a check a direct measurement of $4\pi M_s$ on one of the samples was made using a Faraday balance. This gave a value of 14.7 ± 0.5 kG for Fe-Pd sample C ($d = 22$ Å), in agreement with the value obtained from the Brillouin scattering measurement.

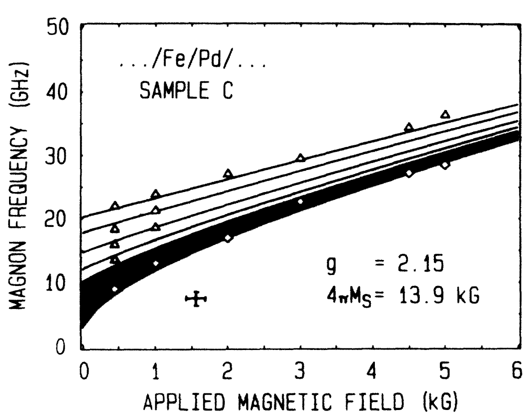

FIG. 2. Experimental frequencies of the lower band edge (◇) and of the discrete modes (△) of sample C (90 bilayers) as a function of applied magnetic field. The solid lines are the calculated mode frequencies (see text), where the dark area contains the experimentally unresolved, remaining 86 modes. The angle of incidence was 43°. A typical error bar is indicated.

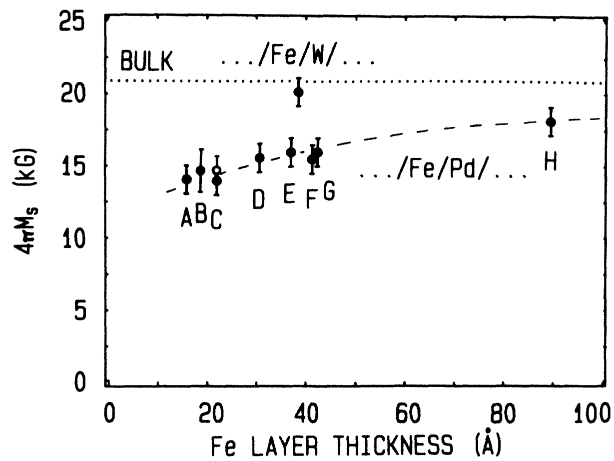

FIG. 3. Fitted values (●) of the saturation magnetization $4\pi M_s$ for Fe-W and Fe-Pd multilayer structures as a function of the Fe layer thickness. For Fe-Pd sample C the value denoted by (○) was measured by a Faraday balance. The dotted line indicates the bulk magnetization of Fe, the dashed line is a guide to the eye through the data of the Fe-Pd multilayer structures.

The strong decrease of $4\pi M_s$ of the Fe-Pd samples, contrary to the Fe-W multilayer structure, may be caused by two mechanisms. First by a charge transfer from Fe to Pd because of the large density of d states near the Fermi level of Pd, and/or second by a strong hybridization between adjacent layers. Because of the different crystalline structures of the nonmagnetic fcc Pd(111) layers and magnetic bcc Fe(110) layers, an additional uniaxial, out-of-plane anisotropy can be induced due to anisotropic stresses, i.e., due to distortions in the crystal angles. The latter effect would be different in the case of bcc W(110), which has the same crystal structure as Fe. Both mechanisms will become increasingly important with decreasing modulation wavelength. Any effect of in-plane anisotropies on $4\pi M_s$ could not be detected; by rotating the samples about the stack axis no variation of the spin-wave spectrum was observed.

An additional reduction of $4\pi M_s$ due to interdiffusion cannot be excluded. Since our model utilizes a mean value of $4\pi M_s$, which is averaged over the spatial distribution in each Fe layer, a strong reduction of $4\pi M_s$ at the interfaces may account for the observed reduction of the averaged value. In fact, at the interfaces the thickness of the zone can be estimated in each Fe layer where ferromagnetism is inhibited by interdiffusion. If the thickness d (d_0) of the magnetic (nonmagnetic) material is changed, characteristic modifications of the field dependence of the calculated spin-wave frequencies result. From the dependence of the mode frequencies in Fig. 2 on d and d_0 the reduction of d, which is followed by a corresponding decrease of $4\pi M_s$, is at most 30%. However, the obtained value of $4\pi M_s$ for sample C agrees with the independently measured value obtained by a Faraday balance (see Fig. 3) only if the reduction of d is less than 10%, i.e., less than the thickness of one atomic layer. It is impossible at present to yield more information about the interdiffusion zone either by light scattering or x-ray diffraction. However, future work studying the different behavior of Fe-Pd and Fe-W multilayer systems will address this question.

In conclusion, we have observed for the first time the shape of the collective spin-wave excitations in multilayer structures with large numbers ($49 \leq N \leq 90$) of bilayers. In agreement with theoretical predictions we found a pronounced asymmetry of the excitation band. Discrete modes are observed in the upper part of the excitation band which are caused by the quantization of $q_\perp$ due to the finite though large number of layers. The surfacelike as well as bulklike character of the excitation band has been demonstrated experimentally. The anomalously strong softening of the saturation magnetization for small modulation wavelengths in the Fe-Pd multilayer systems, contrary to the Fe-W system, may be due to the particular electronic structure of Pd and to its anisotropically distorted crystalline structure.

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