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Oscillatory interlayer exchange coupling of Co/Ru multilayers investigated by Brillouin light scattering

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We have determined the absolute value of the interlayer exchange coupling constant A_{12} of sputtered Co/Ru multilayers using Brillouin light scattering. The spin-wave frequencies, and therefore A_{12}, are found to oscillate as a function of the Ru layer thickness with a period of 11.5 Å. Detailed calculations show that the spin-wave frequency oscillations result from the canting of layer magnetizations in the antiferromagnetic coupling regimes.

Oscillatory, i.e., alternating, ferromagnetic and antiferromagnetic interlayer exchange coupling has become one of the most discussed phenomena in layered magnetic structures in the last few years.[1] Grünberg *et al.*[2] first discussed experimental evidence for antiferromagnetic interlayer exchange coupling in Fe/Cr/Fe sandwich structures, followed by the discovery of oscillatory interlayer exchange as a function of the spacer-material thickness in different multilayered structures by Parkin, More, and Roche.[3] Since then many other systems with antiferromagnetic or oscillatory interlayer coupling have been found,[1,4–7] recently even with two distinct oscillation periods.[8–10] The interest in these phenomena was boosted by the discovery of the so-called "giant magnetoresistance" effect in the antiferromagnetic coupled regimes,[11,12] creating the effect a promising subject for, e.g., designing magnetoresistive pick-up heads for magnetic storage devices.

Multilayered structures of alternating Co and Ru layers show pronounced oscillations of the interlayer exchange coupling constant A_{12}.[3,13] Up to three oscillation cycles were observed[3] by vibrating-sample magnetometry with a periodicity of 12 Å. The saturation fields are larger than 10 kG for Ru thicknesses, d_{Ru}, of smaller than 5.7 Å, and they are in the range of 3.5 kG otherwise. Recently the antiferromagnetic coupling mechanism has been confirmed for Co/Ru multilayers by polarized neutron-scattering measurements.[14]

Oscillations of the interlayer exchange-coupling strength, described by the parameter A_{12}, were observed by magnetometry measurements of the saturation field,[3] although this method can only give access to A_{12} in the antiferromagnetic regimes. For the ferromagnetic regimes spin-engineered structures were investigated.[15] An easier access to A_{12}, both in the ferromagnetic and the antiferromagnetic regimes, is provided by Brillouin light scattering from thermal spin-wave excitations.[2,16,17]

Brillouin light scattering from spin-wave excitations has been used to determine A_{12} both in the ferromagnetic and the antiferromagnetic coupling regimes of sandwich structures consisting of two magnetic layers with a nonmagnetic spacer layer in between.[17] Two spin-wave modes exist,[2] one being a uniform mode across the sandwich structure or an in-phase spin precession of the two layers and the other being the "optic" mode for which the spin precession is out of phase between both magnetic layers. The frequency of the latter mode depends sensitively on the interlayer exchange coupling. Hence a frequency measurement of this mode allows us to determine A_{12}. So far Brillouin light scattering has not yet been applied to antiferromagnetic coupled multilayered structures. The rather complicated spin-wave mode spectrum, as well as the lack of a suitable theoretical model for spin-wave frequency calculations in these structures, taking both interlayer exchange anisotropy, as well as the canting between magnetization directions in different magnetic layers in the antiferromagnetic coupled regimes into account, has hampered this kind of investigations up to now.

In multilayered structures, consisting of alternating magnetic and nonmagnetic layers, dipolar spin-wave modes exist within each magnetic layer (so-called Damon-Eshbach modes), which couple across the intervening nonmagnetic layers. Due to the coupling, which is dipolar and may contain exchange contributions as well, the spin-wave modes form a band of collective spin-wave excitations. One of these modes, the so-called stack surface mode, has its mode energy localized near one of the stack surfaces, and the spins of all layers precess in phase.[18,19] For the latter reason, the frequency of this mode does not depend on the interlayer exchange-coupling strength; however, it is sensitive to the net magnetization of the multilayer stack (canting angle). The spin-wave frequencies of all other modes depend on both the interlayer exchange constant as well as the layer-to-layer distribution of the directions of the magnetization.[18–20]

In the regime of large antiferromagnetic interlayer exchange coupling, neighboring magnetic layers are aligned predominantly antiparallel. Here a new collective spin-wave mode occurs, which is reminiscent of the "optic" high-frequency spin-wave mode of antiferromagnetic bulk material.[19] This mode goes soft with decreasing canting angle between neighboring magnetic layers.

The Co/Ru superlattice structures were deposited on chemically etched Si(111) wafers in a high-vacuum dc magnetron sputtering system containing four magnetron sources. The base pressure of the vacuum system prior to deposition was better than 2×10^{-9} Torr. The structures were prepared in 3.2 mTorr of argon at a deposition rate of 2 Å/s at room temperature. All samples were prepared in one batch without breaking the vacuum via computerized control of the substrate platform and shutters located between each magnetron source and the platform.

The Brillouin light-scattering measurements were performed using a computer-controlled (3+3) pass tandem Fabry Perot interferometer with spectral ranges chosen between 10 and 100 GHz. The incident laser light (514.5-nm Ar^+ line) was focused onto the surface of the sample with a power of up to 200 mW. The scattering angle of the incident laser light was chosen between 20° and 60°. The direction of the spin-wave propagation determined by the scattering geometry was aligned perpendicular to the applied magnetic field. The latter was applied parallel to the layer planes and the backscattered light was detected by a photomultiplier in the depolarized configuration. Due to the weak magnetic light-scattering cross section, data accumulation times of up to 20 h per spectrum were used.

Figure 1 shows four spectra of Co/Ru multilayers with a Co layer thickness of 20 Å and a Ru layer thickness of (a) 20.9 Å, (b) 15.2 Å, (c) 9.5 Å, and (d) 3.8 Å measured with an applied magnetic field of 1 kG. The peaks at ±8.5 GHz correspond to the surface phonon (Rayleigh mode) of the system. Although these signals are largely suppressed by the depolarized configuration, the long accumulation time still results in a considerable contribution. In all spectra we observe a band of collective spin-wave excitations in the frequency range between 10 and 20 GHz. Near 19 GHz the stack surface mode (marked in Fig. 1 with an open arrow) is identified in Figs. 1(a) and 1(c) by its characteristic Stokes/anti-Stokes intensity asymmetry.[21] This pronounced mode is only observable in the regimes of $d_{\mathrm{Ru}}=10$–14 and 20–24 Å, which we identify as the ferromagnetic coupled regimes. Otherwise [cf. Figs. 1(b) and 1(d)] the mode is shifted to lower frequencies and merges with the other band modes.

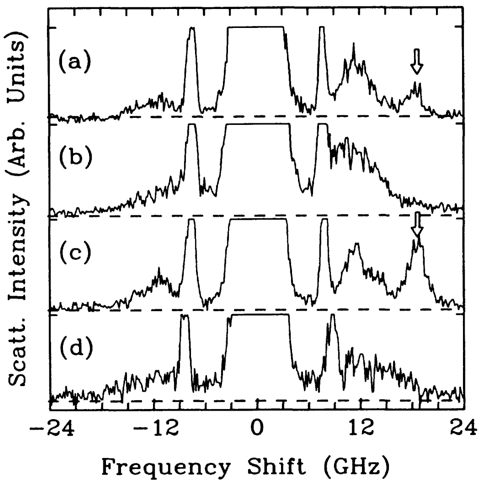

FIG. 1. Spin-wave spectra of Co/Ru multilayers with a Co layer thickness of 20 Å and a Ru thickness of (a) 20.9 Å, (b) 15.2 Å, (c) 9.5 Å, and (d) 3.8 Å. The magnetic field applied perpendicular to the spin-wave propagation direction is 1 kG. The angle of light incidence is 45°. The background due to the photomultiplier dark count rate is indicated by dashed lines.

In the upper part of Fig. 2 the frequency positions of the stack surface mode (squares) and the center of the bulk modes (circles), measured at an applied field of 3 kG, are plotted as a function of the Ru layer thickness. Oscillations with a period of 11.5 Å are well resolved. For comparison, the spin-wave frequencies have been calculated for the exchange-uncoupled case ($A_{12}=0$) using a model described elsewhere.[16] They are shown as full lines in Fig. 2, upper part. The frequencies are adjusted to the experimental data of the stack surface mode in the ferromagnetic coupling regimes by choosing an appropriate value for the uniaxial perpendicular anisotropy constant of 4.7×10^6 erg/cm^3 typical for Co layers. The regimes of Ru thickness exhibiting reduced spin-wave frequencies are identified as the antiferromagnetic coupling regimes, as described further below. In particular, for $d_{\mathrm{Ru}}<5.7$ Å a large antiferromagnetic coupling consistent with earlier observations[3] is found by the large fre-

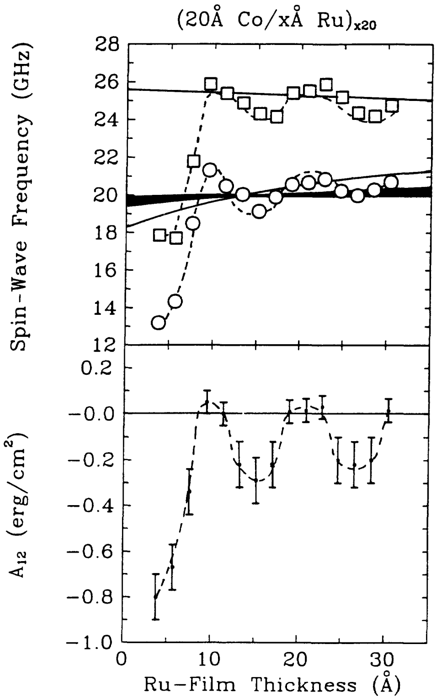

FIG. 2. Upper part: Spin-wave frequencies of the stack surface mode (open squares) and the center of the bulk modes (open circles) as a function of the Ru layer thickness measured at an applied field of 3 kG. For comparison, the spin-wave frequencies calculated for zero interlayer exchange coupling are shown as full lines. Lower part: Determined values of the interlayer exchange constant, A_{12}, as a function of the Ru layer thickness.

quency decrease. The observed frequency oscillations as a function of d_{Ru} are accompanied by oscillations with the same periodicity in the linewidths of the spin wave modes.

We will now discuss the spin-wave properties in the antiferromagnetic coupling regimes. For not too large external fields the magnetizations of neighboring layers are canted with respect to each other. The canting angle depends on the (negative) value of A_{12} and the strength of the applied field. Our calculations are based on an effective medium model, described elsewhere.[18,19] We treat the total multilayer stack as a ferromagnetic film with effective susceptibilities. The susceptibilities are calculated assuming that the electromagnetic fields vary only slightly across each period. Therefore this model is strictly applicable only to the calculation of the stack surface mode and the first few bulk modes. In Fig. 3 we have plotted for Co/Ru multilayers the spin-wave frequencies of the surface mode and of the lowest order bulk mode as a function of the interlayer exchange constant A_{12}. The Co and Ru thicknesses are both 20 Å, and a uniaxial perpendicular anisotropy of 4.7×10^6 erg/cm^3 has been assumed. The applied field is 3 kG. For comparison, the calculated canting angle of the saturation magnetization between neighboring magnetic layers (see below) is shown as a dashed line. For $A_{12} > -0.25$ erg/cm^2, i.e., for zero canting angle, the spin-wave frequency of the stack surface mode is independent of A_{12}, since here the net magnetization is constant.

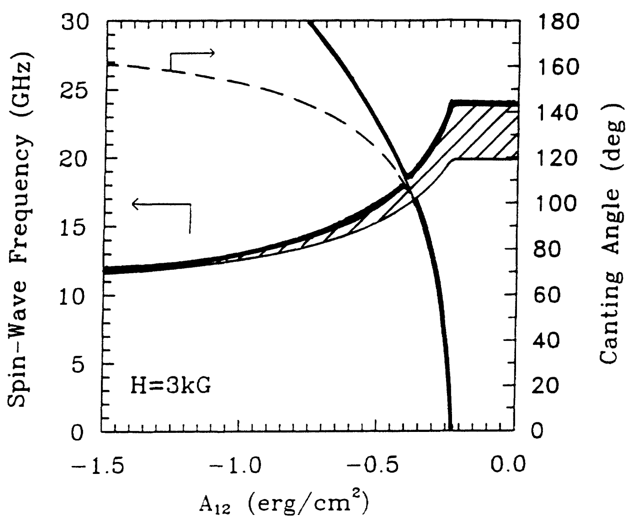

FIG. 3. Spin-wave frequencies calculated as a function of the interlayer exchange constant, A_{12}, using the effective-medium approach described in the text. The Co and Ru layer thicknesses are both 20 Å. The applied field is 3 kG; the uniaxial anisotropy is 4.7×10^6 erg/cm^2. The canting angle between the saturation magnetizations of neighboring layers is shown as a dashed line.

Canting of the magnetization occurs in Fig. 3 for $A_{12} < -0.25$ erg/cm^2. Here the spin-wave frequencies display a much more complicated behavior. There are now two surface modes, indicated in the figure by thick solid lines, and we see that one of these surface modes goes soft when the magnetizations lie parallel to one another at $A_{12} = -0.25$ erg/cm^2. The bulk spin-wave bands are shown as hatched areas. For $d_{\mathrm{Co}} = d_{\mathrm{Ru}}$, the surface modes are not well defined and exist at the top of the bulk bands, thus forming the upper frequency limit of the dipolar bulk modes.

There are now two bands. One band is the continuation of the collective spin-wave band of ferromagnetically coupled layers. Its frequencies decrease with increasing negative value of A_{12}, i.e., increasing canting angle. This band is crossed by a new collective band, which is reminiscent of the optic high-frequency spin-wave mode of antiferromagnetic bulk materials, and which goes soft for $A_{12} > -0.25$ erg/cm^2. Within this band and apart from the crossing regime, all modes are degenerate on the scale of Fig. 3. The behavior of the former band, apart from the crossing regime, can be easily understood. The modes respond to the net magnetization in the direction of the field. As the canting angle increases the net magnetization in the direction of the field decreases approximately according to

$$\cos\alpha = -\frac{H_0 M}{2A_{12}} , \tag{1}$$

where M is the net magnetic moment of a film per unit area and α is the canting angle.[19] In the simple case of a semi-infinite multilayer without anisotropies, the frequency of the surface mode is well described by

$$\frac{\omega}{\gamma} = H_0 + 2\pi M_s \cos\alpha . \tag{2}$$

Similarly, the bottom of the associated bulk band is given by

$$\frac{\omega}{\gamma} = [H_0(H_0 + 4\pi M_s \cos\alpha)]^{1/2} . \tag{3}$$

Thus measurements of the frequencies are direct measures of the interlayer exchange constant A_{12}. We emphasize that modes at these frequencies make the largest contribution to the light-scattering cross section. For superlattice structures of finite thickness with anisotropies, however, the frequencies can only be determined numerically. Fitting our model to the experimental data, values for the interlayer exchange coupling constant, A_{12}, are obtained. They are displayed in the lower part of Fig. 2. Although the error bars are rather large due to the experimentally observed large linewidth of the modes, the oscillations from ferromagnetic to antiferromagnetic coupling are clearly observed. However, the error bars are too large to determine the decay in oscillation amplitude with increasing d_{Ru}.

Finally, we note that recent work indicates that for certain fields H_0 the ground state of an antiferromagnetically coupled multilayer may be quite different from the state assumed here.[22] Work is currently under way to examine the spin-wave frequencies in these cases.

In summary, we have investigated the ferromagnetic and antiferromagnetic coupling of Co/Ru multilayers by Brillouin light scattering. We conclude by commenting on the obtained values for the interlayer exchange-coupling constant A_{12}. Using magnetometry, A_{12} is essentially obtained from the measured *energy change* from antiparallel to parallel alignment enforced by an applied field. On the other hand, in Brillouin light scattering the spin-wave frequencies are sensitive to the magnetic *torque* between neighboring magnetic layers, mediated by the coupling mechanism, and evidenced by the magnetization canting. In contrast to sandwich structures, a variation in canting directions might be possible in a multilayer structure, and the value of the canting angle would thus vary about a mean value, which is smaller than that of sandwich structures. However, we do not observe such an effect on A_{12} in the canting region: A measurement of the saturation field using a superconducting quantum interference device magnetometer yields for the first antiferromagnetic oscillation ($d_{\mathrm{Ru}}=3.8$ Å) a value for A_{12} of (-0.81 ± 0.04) erg/cm^2, which agrees excellently with the value of (-0.80 ± 0.10) erg/cm^2 obtained from Brillouin light scattering.

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