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# Observation of the exchange-coupled modes in Fe(001)/Pd/Fe(001) ultrathin trilayers

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Trilayers consisting of two films of bcc Fe(001), 5 and 10 ML thick, and separated by 4, 5, 5.5, 6, 8, 10 and 11 ML of Pd were prepared on an Ag(001) substrate by means of molecular beam epitaxy. The magnetic properties of these films were measured using ferromagnetic resonance and Brillouin light scattering techniques. The exchange coupling between the two iron films was found to be very strong for four or less ML of Pd. This exchange coupling was observed to become very weak for Pd films 5 or more ML thick ($|J| \leq 0.1\,\text{erg/cm}^2$).

We have used room temperature ferromagnetic resonance (FMR) and Brillouin light scattering (BLS) experiments to study the coupling between two ultrathin films of bcc Fe(001) separated by a few monolayers of Pd. Trilayer films were prepared in which 5 monolayers (ML) of Fe(001) were grown layer-by-layer on a suitably prepared Ag(001) substrate, a variable number of ML of Pd were grown layer-by-layer on the Fe(001) film, and a further 10 ML of Fe(001) were grown on the Pd film. This trilayer structure was capped by a further deposition of 20 ML of Au(001) in order to protect the ultrathin Fe and Pd layers when the specimens were removed from the vacuum system in order to carry out the FMR and BLS measurements. Films were prepared having Pd thicknesses of 4, 5, 5.5, 6, 8, 10 and 11 ML. The ultrathin layers of Fe and Pd were prepared in ultrahigh vacuum ($\approx 10^{-10}$ Torr) using the techniques of molecular beam epitaxy (MBE). The substrate was held at room temperature during deposition of the films. Specific details of the growth of Pd on Fe(001) and of the growth of Fe(001) on Pd will be described elsewhere together with a report on the temperature dependence of their magnetic properties [1]. The layer-by-layer growth of Fe(001) on singular Ag(001) surfaces has been described in previous publications [2, 3].

FMR measurements at 36.3 and 73 GHz were carried out using cylindrical $TE_{10}$ resonant cavities and standard magnetic field modulation techniques to obtain the derivative of the absorption with respect to field [2]. The BLS measurements were carried out by means of a Sandercock 4 + 2 pass tandem Fabry–Perot interferometer [4]. The two lowest normal mode frequencies for a pair of coupled magnetic films correspond to an acoustic mode, in which the coupled magnetizations precess in phase, and an optical mode in which the magnetizations precess out of phase [2]. The nature of these modes has also been discussed by Grünberg [5] for identical films. In an FMR experiment one therefore expects to observe two magnetic fields at which resonant absorption takes place for a given frequency, providing that the coupling between the two Fe films is not too strong: one resonance corresponds to absorption by the acoustic mode and the other to absorption by the optical mode. These two absorption lines were observed for all Fe/Pd/Fe trilayers for which the thickness of the Pd interlayer was 5 ML or greater. Similarly, two pairs of Stokes–anti-Stokes lines were observed in BLS for a given applied magnetic field corresponding to the

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acoustic and optical modes (see fig. 1). FMR and BLS data obtained for the trilayers were analyzed using a calculation based on the Landau–Lifshitz equations of motion for the magnetization combined with the Rado–Weertman [6] boundary conditions and a magnetic exchange interaction between two magnetic films of the form used by Hoffman et al. [7],

$$E^{\mathrm{AB}} = J\left(\frac{\boldsymbol{M}^{\mathrm{A}} \cdot \boldsymbol{M}^{\mathrm{B}}}{M_{\mathrm{A}} M_{\mathrm{B}}}\right) \mathrm{erg/cm^2}. \tag{1}$$

$\boldsymbol{M}^{\mathrm{A}}, \boldsymbol{M}^{\mathrm{B}}$ are magnetization densities which, in films as thin as those used in this work, are uniform across the thickness of each magnetic layer. $M_{\mathrm{A}}, M_{\mathrm{B}}$ are the magnitudes of the magnetization density in the two Fe films. Positive $J$ corresponds to antiferromagnetic coupling and negative $J$ corresponds to ferromagnetic coupling between the films. In our experiments the two Fe films were found to be coupled ferromagnetically for all Pd thicknesses. This simplified the theoretical treatment enormously since the equilibrium magnetizations could be assumed to be parallel in the two films. The theory for the case of FMR (driven magnetizations) has been discussed by Heinrich [2] et al., the exchange parameter in their work has been written in the form

$$J = 4(A^{\mathrm{AB}}/a), \tag{2}$$

where $a = 2.86$ Å is the bcc cube edge in iron and where $A^{\mathrm{AB}}$ is an exchange stiffness parameter which may be expected to vary between a value comparable to the bulk exchange stiffness ($\approx 10^{-6}$ erg/cm) for strongly coupled films and zero for the limit of completely uncoupled films. The magnetic field dependencies of the frequencies and intensities of the scattered light measured in the BLS experiment were compared with the calculations described by Cochran and Dutcher [8, 9]. It can be shown that the optical mode for two films having identical magnetic properties, but not necessarily the same thick-

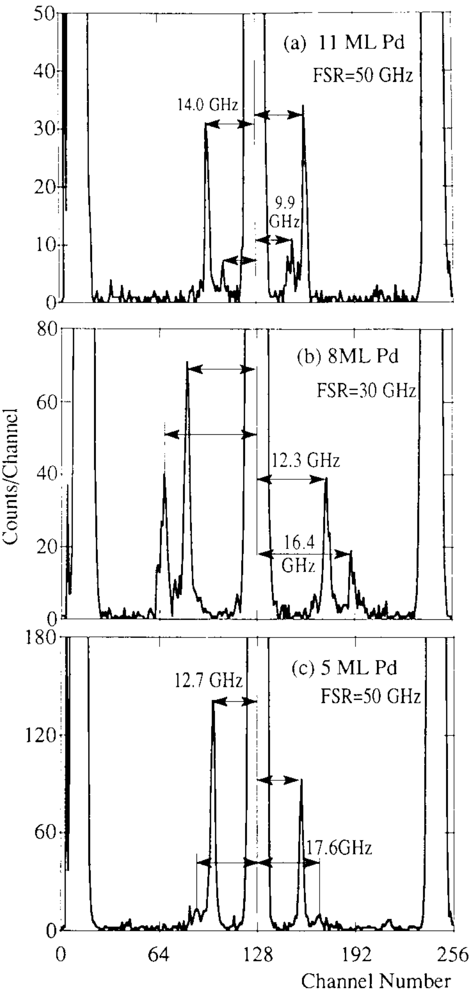

Fig. 1. Scattered light intensity vs. frequency shift at a magnetic field of 0.82 kOe applied along an easy axis for three different Fe/Pd/Fe trilayers (see table 1). The 5145 Å light was incident at 45° and collected in the back-scattering configuration; the in-plane component of the optical wave-vector was $1.73 \times 10^5\ \mathrm{cm}^{-1}$. (a) 11 ML of Pd. The two iron films are very weakly exchange coupled ($J \approx 0$). The most intense peak is due to the acoustic mode and is shifted by a greater frequency than the weaker peak which is due to the optical mode. The mode splitting in this case is primarily caused by the uniaxial surface energy density of eq. (4); there is, however, a weak magnetostatic coupling between the two iron films (antiferromagnetic) due to the in-plane component of the optical wave-vector. (b) 8 ML of Pd. The exchange and magnetostatic coupling are comparable in magnitude ($J = -0.08\ \mathrm{erg/cm^2}$) but the exchange coupling exceeds the magnetostatic coupling. The most intense peak, corresponding to the acoustic mode, is now lower in frequency than the peak corresponding to the optical mode. (c) 5.0 ML of Pd. The exchange coupling exceeds that for 8 ML of Pd ($J = -0.12\ \mathrm{erg/cm^2}$).

nesses, exhibits nearly zero microwave absorption except for a very narrow range of the coupling parameter, $J$: this neglects the spatial variation of the microwave driving field across the films due to the microwave skin effect. The microwave skin-depth, even at FMR, is $\approx 10^3$ Å and is therefore very large compared with the film thicknesses used in our experiments. Similarly, neglecting the small difference in optical electric field amplitude within the two feromagnetic films, the scattered light amplitudes from the two films corresponding to the optical mode of the coupled system cancel if the lowest normal mode frequency in each of the isolated films is the same. It is for that reason that we have prepared trilayer structures in which the lowest modes for each of the isolated layers for a given applied magnetic field are quite different. This has been achieved by using Fe films having different thicknesses (5 and 10 ML). The difference in frequency between the two films is a consequence of a uniaxial surface energy contribution to the effective magnetization:

$$4\pi M_{\text{eff}} = 4\pi M_s - (2K_u/dM_s), \qquad (3)$$

where $d$ is the film thickness, $M_s$ is the saturation magnetization density, and $K_u$ is a parameter which specifies a magnetic surface energy density of the form

$$E_s = -K_u(M_z/M_s)^2 \text{ erg/cm}^2, \qquad (4)$$

$M_z$ is the magnetization component along the film normal. The light scattering intensity corresponding to the optical mode is always smaller than that corresponding to the acoustic mode because the radiation scattered from the individual iron films interfere destructively when their magnetizations precess out of phase. It is worth noting that for the BLS experiment and for zero interlayer exchange coupling the lowest frequency corresponds to the optical mode (see fig. 1). This is a consequence of magnetic dipole coupling between the two films which is antiferromagnetic: see Grünberg [4] section 8.3.2. As the ferromagnetic exchange coupling increases the frequency of the optical mode increases: for our films the optical mode frequency is expected to exceed the acoustic mode frequency for $|J|$ greater than $\approx 0.04$ erg/cm$^2$.

FMR data for 36.3 and 73 GHz were used to deduce values for the $g$-factors, effective magnetizations, and 4-fold in-plane magnetocrystalline anisotropy parameters for the single 5 and 10 ML Fe films [10] in contact with 8 ML of Pd. These parameters were used as the initial values for fitting FMR data at 36.3 GHz obtained for the

Table 1

A comparison of exchange coupling parameter, $J$, and uniaxial surface anisotropy parameters, $K_{\text{UA}}$, $K_{\text{UB}}$, for room temperature Fe/Pd/Fe trilayers obtained from ferromagnetic resonance experiments at 36.3 GHz (FMR) and from Brillouin light scattering data (BLS) using back-scattered 5145 Å light incident at 45°. $J$ is defined by eq. (1) and $K_{\text{UA}}$, $K_{\text{UB}}$ by eq. (4) of the text. Fe(001) layer A was 10 monolayers (ML) thick and Fe(001) layer B was 5 ML thick: the thickness of the intervening Pd layer is indicated in column 1. $K_{1\text{A}}$ and $K_{1\text{B}}$ are 4-fold in-plane magnetocrystalline anisotropy constants determined from the FMR measurements (in units of $10^5$ erg/cm$^3$). Other parameters used for film A (film B) were: saturation magnetization[a] = 20.04 (21.55) kOe; $g$-factor = 2.08 (2.10)

| Pd (ML) | $K_{1\text{A}}$ | $K_{1\text{B}}$ | $-J$ (erg/cm$^2$) |  | $K_{\text{UA}}$ (erg/cm$^2$) |  | $K_{\text{UB}}$ (erg/cm$^2$) |  |
|---|---|---|---|---|---|---|---|---|
|  |  |  | FMR | BLS | FMR | BLS | FMR | BLS |
| 5 | 2.05 | 1.01 | 0.115 | 0.115 | 0.53 | 0.50 | 0.71 | 0.72 |
| 5.5 | 1.68 | 1.30 | 0.073 | 0.063 | 0.53 | 0.25 | 0.71 | 0.68 |
| 6 | 1.67 | 1.04 | 0.044 | 0.044 | 0.53 | 0.53 | 0.75 | 0.80 |
| 8 | 1.57 | 0.99 | 0.080 | 0.08 | 0.53 | 0.30 | 0.75 | 0.72 |
| 10 | 1.91 | 0.97 | 0.028 | – | 0.54 | – | 0.77 | – |
| 11 | 1.82 | 0.94 | 0.027 | 0.00 | 0.55 | 0.40 | 0.74 | 0.74 |

[a] Obtained from FMR absorption strength measurements [3] on the single Fe–Pd films (see the text). These magnetizations have little effect on the values of $J$ used to fit the data; they do affect the surface energy parameters $K_{\text{UA}}$ and $K_{\text{UB}}$.

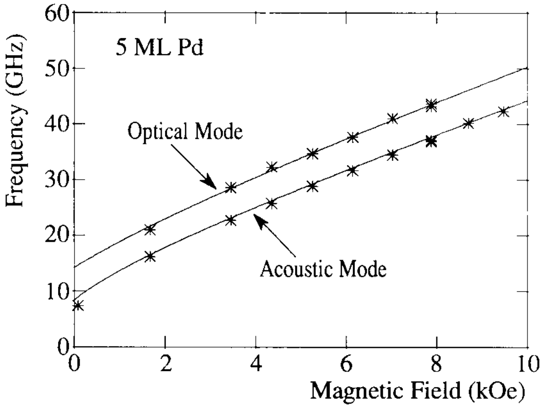

Fig. 2. Magnon frequency vs. applied magnetic field for the trilayer (Ag(001)/5 Fe/5 Pd/10 Fe/20 Au), see table 1, measured by means of Brillouin light scattering. The field was applied in the plane of the film and along a cube axis (easy axis). The solid lines were calculated [8, 9] using the BLS parameters listed in table 1.

tri-layer films. The $g$-factors were fixed at the values $g_A = 2.08$ and $g_B = 2.10$ corresponding to the single films; the effective magnetizations, the in-plane anisotropies, and the coupling parameter, $J$, were varied to obtain the optimum least squares fit. The results of this procedure are listed under FMR in table 1. The parameters which were used to fit the FMR data were used to compare calculated and observed magnetic field dependencies of the mode frequencies measured by means of BLS. These magnetic parameters were then adjusted in order to obtain a satisfactory agreement between the BLS data and the calculated curves. An example of the degree of agreement between calculated and observed BLS frequencies is shown in fig. 2. For all trilayers for which the Pd layer was thicker than 4 ML the model of exchange coupled magnetic layers based on eq. (1) provided a satisfactory description of the data. The agreement between magnetic parameters deduced from FMR and from BLS was satisfactory (see table 1). It must be borne in mind that the FMR absorption is averaged over the entire specimen whose diameter is ≈ 1 cm, whereas the BLS measurement probes an area whose diameter is that of the focussed laser beam, $\approx 2 \times 10^{-3}$ cm.

The trilayer fabricated with a 4 ML Pd interlayer (Ag(001)/5 Fe/4 Pd/9.3 Fe/20 Au) proved to be a special case. This specimen exhibited only a single FMR absorption line for a given frequency, and the BLS measurements revealed only a single pair of Stokes–anti-Stokes lines. The specimen behaved like a single ferromagnetic film; the strength of the FMR absorption indicated that the Pd in this trilayer very likely became ferromagnetic. Perhaps the most remarkable result of this investigation has been this observation of a very sudden transition from the strong coupling limit to the weak coupling limit as the thickness of Pd was increased from 4 to 5 ML.

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[10] For the 5 Fe/8 Pd film $4\pi M_{\text{eff}} = 9.40$ kOe, $g = 2.10$, $K_1 = 1.01 \times 10^5$ erg/cm$^3$. For the 8 Pd/10 Fe film $4\pi M_{\text{eff}} = 16.94$ kOe, $g = 2.08$, $K_1 = 1.49 \times 10^5$ erg/cm$^3$.