X-ray Detected Ferromagnetic Resonance of an Exchange Coupled NiFe/CoFe Bilayer

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Abstract -- Strongly exchanged coupled hard and soft magnetic layers find extensive use in data storage applications. However, it has so far been difficult to access the behavior and precise influence of each individual layer. The synchrotron radiation-based technique of x-ray detected ferromagnetic resonance (XFMR) allows element-specific and phase-resolved ferromagnetic resonance (FMR) measurements in the frequency range from 0.5 to 10 GHz. The magnetization dynamics of a strongly exchange coupled NiFe(40 nm)/CoFe(40 nm) bilayer was studied by detecting the FMR for each layer in the film using x-ray magnetic circular dichroism at the L₂ absorption edges of Ni and Co. The epitaxially grown bilayer exhibits two principal resonant modes, involving both layers simultaneously. These modes can be classified as a NiFe dominated acoustic mode and a CoFe dominated optical mode. In the acoustic mode the Co and Ni spin precessions are in phase, whereas in the optical mode the phase difference was found to be ~166°. Using the Kittel resonance equation, it is found that the ‘strongly coupled spin-modes’ differ from the acoustic and optical excitations present in bilayers systems with non-magnetic spacers. This is due to a transfer of the anisotropy from the hard CoFe to the softer NiFe layer via the strong exchange coupling. This conclusion is supported by micromagnetic simulations. The results provide a comprehensive picture of the dynamics of magnetic multilayers.

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I. Introduction

In recent years, multilayers consisting of alternating hard and soft magnetic layers have attracted a great deal of attention due to their potential applications in data storage media [1-3], permanent magnets [4-5], and microelectromechanical systems [6-8]. In particular, bilayer systems composed of exchange-coupled hard and soft layers have been proposed as superior data storage media because they can overcome the so-called superparamagnetic limit while retaining relatively low writing fields. Further, exchange coupling between layers should lead to increased damping (see also 9) – and, due to higher resonance frequencies, – to faster writing speeds. For such applications [10] it is

Magnetism and Superconductivity
Gavin Stenning
XFMR Paper
important to gain detailed knowledge of the bilayer magnetization dynamics. Here we focus on the study of a bilayer system consisting of a magnetically soft NiFe layer, exchange-coupled to a harder CoFe layer. Such systems are commonly exploited in nonreciprocal microwave and spin torque transfer devices [11].

Ferromagnetic resonance (FMR) is widely used to study magnetization dynamics of multilayer systems [12-15] and exchange-coupled bi- and trilayer systems [16]. Both broadband coplanar waveguide (CPW) and resonant cavity FMR techniques have been commonly employed. However, these methods are limited in that they can only detect the net response of the multilayer sample. In practice, the response of individual layers can only be inferred indirectly by comparing the experimental FMR spectrum with modeled spectra [14]. X-ray detected FMR (XFMR), on the other hand, is a powerful synchrotron radiation based-tool which can be used to study element-selective magnetization dynamics [10, 11, 17-21]. Magnetic and chemical contrast is provided by x-ray magnetic circular dichroism (XMCD) [22], while the phase difference of the spin rotations can be determined using stroboscopic XMCD measurements. Element specificity leads to clear advantages of XFMR over FMR, e.g., when two layers are magnetically identical, the intensity of the optical mode in FMR will be zero. In detail, the intensity of the optical mode is proportional to the difference in effective magnetization of the two layers and inversely proportional to the interlayer exchange [23]. However, this problem is negated in XFMR where, element specificity can be used to isolate the response of the individual layer, provided they have different composition.

Here, we use XFMR to investigate a NiFe (40 nm)/CoFe (40 nm) bilayer by tuning the photon energy to the Ni and Co L_{2,3} x-ray absorption edges. The XFMR study is complemented by vector network analyzer (VNA) based FMR as well as micromagnetic simulations using the simulation code Nmag [24, 25]. It will be shown that the combined approach provides a comprehensive picture of the dynamics of an epitaxial NiFe/CoFe bilayers, compared to weakly coupled systems of ferromagnetic layers separated by a non-magnetic spacer [26].

II. Experimental Details

The magnetic bilayers were grown by molecular beam epitaxy (MBE) on in-situ annealed MgO (001) substrates. After depositing Fe (3 nm) seed and Pt (30 nm) buffer layers, and an annealing step at 500°C for 1800 s, the Ni_{0.81}Fe_{0.19} (40 nm)/Co_{0.5}Fe_{0.5} (40 nm) bilayers were grown by co-deposition at a substrate temperature of 100°C. Finally, to prevent oxidation, the sample was capped with Pt (5 nm) at room temperature. The MBE grown films have atomically abrupt interfaces and well-defined anisotropy. The most significant feature is the four-fold anisotropy of the CoFe film, which induces a transferred anisotropy into the NiFe layer, similar to that witnessed in a CoFe/Cr/NiFe trilayer system [14].

Prior to the XFMR experiments, the sample was characterized using VNA-FMR. To achieve good coupling the bilayer sample was placed top-down directly onto a CPW (“flip-chip” method) [27] and then positioned in an electromagnet. A VNA (HP E5071C) was used to determine the scattering parameter S_{21} of the system. In particular, 2D maps of S_{21} were obtained as a function of frequency and applied field [28]. This allows the net resonant modes of the bilayer system to be identified.

Magnetism and Superconductivity
Gavin Stenning
XFMR Paper
For the XFMR experiments, 100 nm of Cu was deposited onto the bilayer structure and subsequently patterned into a CPW with a characteristic impedance of 50 Ω. The CPW arrangement favors in-phase excitation of the NiFe and CoFe layers [10], while providing maximum microwave driving fields to the underlying NiFe/CoFe film ($B_{rf} = 175 \mu T$) [21].

XFMR was performed both on beamline I06 at Diamond Light Source and on beamline PM3 at BESSY II. A schematic of the experimental setup is displayed in Fig. 1 (see also [9,11,21,29] for alternative approaches). Phase sensitivity in XFMR is achieved by driving the magnetic resonance at harmonics of the electron bunch frequency of the storage ring. The higher harmonics are generated using a custom microwave comb generator driven by the master oscillator of the storage ring, which is ~500 MHz both at Diamond and at BESSY. Filters and amplifiers are used to excite a narrow band, high power (25-30 dBm) radio frequency (rf) field. A delay line enables phase shifting of the microwave oscillation with respect to the x-ray pulses in steps of 0.5 ps. The rotation of the magnetization is detected stroboscopically by measuring the XMCD of the magnetization component rotating normal to the static magnetic field. A limiting factor determining the maximum frequency (14 GHz) detectable by XFMR is the pulse width of the x-rays (~35 ps). However, other factors, such as the microwave losses, may reduce the maximum frequency still further. Timing jitter of the master oscillator signal at Diamond was measured using a spectrum analyzer to be below 0.5 ps.

Phase-resolved XFMR requires the sample to be magnetized normal to the incident x-ray beam, while static XMCD has a maximum signal when the magnetization component is along the x-ray beam. In practice, compromises have to be made and the set-up used for XFMR is shown schematically in Fig. 1, here it will be observed that the x-ray beam makes an angle of 30° with respect to the plane of the sample. At Diamond the applied field was supplied by the portable octopole magnet system (POMS), which allows magnetic fields to be applied in any direction up to 0.9 T. At BESSY the magnetic field was supplied by two sets of orthogonal coils mounted inside the measurement chamber. Incoming x-rays were incident only on the central part of the CPW signal line patterned on the film, therefore avoiding non-uniform rf excitation [14]. The sample plane was oriented at a small angle of incidence to ensure that XMCD is sensitive to the larger in-plane component of the magnetization precession. The x-ray absorption was measured in transmission geometry by detecting the x-ray excited luminescence emerging from the MgO substrate, by using a photodiode which was mounted directly behind the sample.
XFMR was detected by setting the x-ray energy to a resonant absorption edge, the applied field to the appropriate value for the selected rf frequency, and subsequently shifting the rf phase with respect to the x-ray pulses. This enables the precession of the spins to be mapped out with a phase resolution of $\Delta \phi \approx 7^\circ$ at 10 GHz [20]. To double the effective size of the XMCD signal, arising from the small angle precession, the phase of the rf field was modulated between $0^\circ$ and $180^\circ$ for each angle $\phi$ on the precessional cycle and using lock-in detection techniques. Alternatively, XFMR can be measured by sweeping the bias field at a fixed microwave phase with respect to the x-ray pulses. For each point in an XFMR scan, the XAS signal below the resonance absorption energy is subtracted from that at the resonance to yield a relative XMCD amplitude signal (independent of x-ray beam intensity).

III. Characterization of Hybrid Modes

The resonance frequency as a function of applied field, obtained by VNA-FMR, is shown in Fig. 2. Two principal resonances are observed, which we will refer to as the acoustic and optical branch at lower and higher frequencies, respectively, in line with observations for identical ferromagnetic layers coupled via a non-magnetic spacer [26,30,31]. In the Magnetism and Superconductivity
Gavin Stenning
XFMR Paper
acoustic mode, all the spins in both layers are expected to rotate in phase, while in the optical mode, the spins in the two magnetic layers would rotate out-of-phase with respect to each other. The full width half maximum (FWHM) (linewidth) of the resonances at 15 GHz in Fig. 2 are 17.1 mT and 61.7 mT for the acoustic and optical mode, respectively.

FIG 2: (color online) Resonance condition determined by VNA-FMR (symbols). The solid lines are obtained from a least squares fit to the Kittel formula in Eq. (1).

XMFR was subsequently performed to probe the nature of the two resonant modes in more detail. However before such experiments can be performed, it is necessary to determine the Co and Ni $L_{2,3}$ absorption edges. Static XMCD spectra with the magnetic field applied along the x-ray beam, measured in transmission, are presented in Fig. 3. The spectra of both Co and Ni at the $L_3$ edge exhibit saturation effects, because the soft x-rays are attenuated exponentially on passing through the film, which is thicker than the x-ray attenuation length. To reduce the saturation effects in the XFMR experiments the x-ray energies were set to the energies of the $L_2$ edges, marked by the arrows in Fig. 3. The cone angle of precession is of the order of a few degrees [32], so that the dynamic XMCD is about 50 times smaller than its static counterpart.
FIG 3: (color online) Typical static XMCD spectrum for (a) Co and (b) Ni $L_3$ and $L_2$ absorption edges measured in transmission mode. The XFMR was measured at the $L_2$ edges marked by the arrows.

The XFMR precession modes were obtained stroboscopically, by measuring the XMCD signal at $L_2$ as a function of the delay between the rf oscillations and x-ray bunches. The results obtained at 9.5 GHz are shown in Fig. 4.
Magnetism and Superconductivity

Gavin Stenning

XFMR Paper

FIG 4: (color online) Delay scans showing the precession of the Co and Ni spins in (a) the acoustic mode at 71 mT and (b) the optical mode at 6.2 mT. Both scans were obtained at 9.5 GHz. The solid lines represent sinusoidal fits to the data. For the optical (acoustic) mode the phase difference was found to be 166° (4°), respectively.

From Fig. 4 it is evident that the layer averages of the Co and Ni signals are in anti-phase for the optical mode, while they are in phase for the acoustic mode. This justifies our choice of nomenclature for the two modes. More information can be obtained from the relative strengths of the Co and Ni XFMR signals. In the optical mode the amplitude of the Co signal is much larger than that of the Ni, while the opposite is true for the acoustic mode. It can therefore be concluded that the optical mode appears to be driven primarily by the CoFe layer [Fig. 4(b)], while the acoustic mode is dominated by the NiFe layer [Fig. 4(a)]. Similar effects have been observed in resonant trilayer spin valves [32]. This demonstrates that the optical mode, mainly driven by the CoFe, possesses a broader linewidth due to intrinsic losses and damping arising from the anisotropy, which due to exchange coupling induces a broader linewidth in the NiFe than otherwise found in the constituent layer by itself [32].

By accumulating delay scans of the form shown in Fig. 4 an informative picture of the resonant modes can be built up as a function of applied field. (compare also Fig. 4 of Ref. [21] and Fig. 3 of Ref. [32], respectively). In Fig. 5 we show the amplitude and relative phase of the Ni and Co signals as a function of applied field. Near $B_a = 0$ the optical mode is large and the Ni and Co layers are $\approx 180^\circ$ out of phase with each other. As the field is increased, the acoustic mode is reached at 35 mT which is dominated by the Ni signal. In addition the Ni changes phase from $\approx 180^\circ$ to $\approx 0^\circ$ on passing through the Magnetism and Superconductivity

Gavin Stenning

XFMR Paper
acoustic resonance [29]. The Co which is coupled to the Ni, shows a bipolar line shape, with its inflection point at the maximum of the Ni amplitude. Above the acoustic mode resonance \((B_a > 40 \text{ mT})\) the Ni and Co precessions are in phase.

![Graph showing amplitude and relative phase vs. applied field](image)

**FIG 5:** (color online) (a) Amplitude (photodiode current) and (b) relative phase obtained from XFMR precessional scans at the Ni and Co \(L_2\) edges at 8 GHz, as a function of the applied field.

**IV. Micromagnetic Modeling**

To simulate the bilayer dynamics, a quasi-1D model system was investigated using the finite-element micromagnetic simulation code Nmag [24,25]. This code has been previously used to model the dynamics of exchange spring coupled systems [25,33]. The model takes the form of an 80 × 10 × 10 nm\(^3\) cuboid crossing the CoFe/NiFe interface, with the interface in the \(yz\) plane \((x = 0)\), NiFe for \(x < 0\) and CoFe for \(x > 0\) (see Fig. 6). A consequence of using the quasi-1D model was that the demagnetization field had to be ignored to avoid unrealistic alignment perpendicular to the bilayer film. As such this model should be viewed as a qualitative approach giving detailed information about the structure of the various modes but not about the resonance conditions \((i.e.\ frequency vs B_a)\).

The quasi-1D model was prepared in an initial ‘excited state’ by applying a magnetic field \(B_a\) along both the \(y\)-axis and at a small angle \(\theta = 0.6^\circ\) away from the \(y\)-axis \((i.e.\ small\ component\ along\ the\ \(z\)-axis)\). After the magnetization has been allowed to settle into the initial state \(\theta = 0.6^\circ\) and has reached a local equilibrium configuration, the main dynamic simulation then commences. At \(t = 0\) the angle of the applied field is switched from \(\theta = 0.6^\circ\) to \(0^\circ\). Hence, for \(t > 0\), with \(B_a\) along the \(y\)-axis, the system relaxes...
to a new minimum energy configuration. This modest change of the angle of the applied field excites the system sufficiently leading to spin waves propagating through the system. The magnetic response $M(x,y,z,t)$ for $t>0$ was determined using Nmag. A Fourier transform of the spatially resolved and spatially averaged magnetization as a function of time was subsequently performed allowing both amplitude and phase of the modes to be determined. The spatially averaged analysis showed to have little dependence on $z$ and $y$, hence only the $x$ dependence (normal to the film) is shown in Fig. 6.

![Simulated spatial distribution of the (a) acoustic and (b) optical mode, on resonance, across the interface ($x = 0$) of the NiFe/CoFe bilayer.](image)

**FIG 6**: Simulated spatial distribution of the (a) acoustic and (b) optical mode, on resonance, across the interface ($x = 0$) of the NiFe/CoFe bilayer.

The advantage of the 1D model is that it provides insight into the spatial form of each mode observed. Figure 6(a) shows that for the acoustic mode, the signal from the NiFe layer is dominant compared to that of CoFe. In contrast, in Fig. 6(b) it is the signal from the CoFe layer which is dominant in the optical mode. Note also that in Fig. 6(b) the NiFe amplitude varies from positive to negative, therefore leading to a reduction in the net transverse magnetization when averaged over the whole layer. This is in agreement with XFMR data which represents an average over the individual layers.

The XFMR and VNA-FMR data clearly show that the acoustic and optical modes are dominated by the NiFe and CoFe layers respectively and, to a first approximation, can be treated independently. With this in mind a simple approach was used to fit the VNA-
FMR data shown in Fig. 2 by using Kittel’s resonance equation for the two layers taken separately. The Kittel resonance condition for a thin film sample [34] is given by,

\[ \omega_0 = \gamma \sqrt{(B_a + B_k)(B_a + B_k + \mu_0 M_0)} \]

\[ B_k = \frac{2K_1}{\mu_0 M_0} \]

where \( B_k \) and \( K_1 \) are the anisotropy field and anisotropy constant, respectively, while \( M_0 \) is the saturation magnetization. A least-squares fit to the experimental data of Fig. 2 was used to determine the parameters \( M_0 \) and \( B_k \) for each of the layers. For CoFe, \( M_0 \) was found to be \( 1.78 \times 10^6 \) Am\(^{-1} \) and \( K_1 = 46 \times 10^3 \) Jm\(^{-3} \). These values are in close agreement with values found in the literature [35, 36]. By contrast, for NiFe the value of \( M_0 = 9.06 \times 10^5 \) Am\(^{-1} \) [37] was found to be in agreement with previous determinations, while the value of \( K_1 = 12 \times 10^3 \) Jm\(^{-3} \) is much higher than expected. We ascribe the latter to a transfer of residual four-fold anisotropy from the CoFe to the NiFe layer, via the strong exchange interaction across the interface. The results of the least-squares fit to the experimental data are shown in Fig. 2 (solid lines). For the NiFe resonance there is a slight deviation from the fit to the experimental data at low fields. This is due to the presence of an exchange spring [38] in the NiFe layer, pinned at the interface by the anisotropic CoFe. In summary, the bilayer behaves to first order as two separate layers, but with a transfer of the anisotropy from the CoFe to the NiFe layer. The modes predicted by micromagnetic simulations, and by fitting the Kittel formula in Eq. (1), are in reasonable agreement with the experimental data, collected both by VNA-FMR and XFMR.

Finally, the Nmag simulations also provide phase information qualitatively confirming the behavior of the observed modes shown in Fig. 5. For the acoustic NiFe mode, on passing through resonance the phase flips by \( \sim 180^\circ \) in agreement with experimental results. In contrast the non-resonant CoFe layer, does not show phase reversal on passing through the NiFe resonance. This result is consistent with the XFMR data. In summary, the quasi-1D model can be used as a good, first approach to study the dynamics of the bilayer system.

V. Conclusion

Based on VNA-FMR and XFMR studies, and micromagnetic modeling, a detailed understanding of the magnetization dynamics of a strongly exchange-coupled, epitaxially grown NiFe/CoFe bilayer film has been obtained. In particular, XFMR allows the two layers to be probed separately, yielding details of the nature and origin of the two observed resonant modes. The low frequency, high field, mode can be described as an ‘acoustic mode’ with all spins precessing in-phase. For this mode, the amplitude has its’ maximum in the NiFe film, but decays as it penetrates into the CoFe layer. In contrast, the high frequency, low field, mode is an ‘optical-mode’, with the spins of the two layers precessing in anti-phase. In this mode the amplitude is at a maximum in the CoFe film, as demonstrated by XFMR results. Finally, calculations using the simple Kittel equation for FMR indicate that the resonances of the two layers comprising the NiFe/CoFe bilayer film act predominantly in the constituent layer. However, it is also clear that the
anisotropy of the CoFe film has been ‘transferred’ to the softer NiFe film via the strong exchange coupling across the bilayer interface.

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Magnetism and Superconductivity
Gavin Stenning
XFMR Paper
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