

Weakly coupled motion of individual layers in ferromagnetic resonance

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(Received 26 April 2006; published 15 August 2006)

We demonstrate a layer- and time-resolved measurement of ferromagnetic resonance (FMR) in a $\text{Ni}_{81}\text{Fe}_{19}/\text{Cu}/\text{Co}_{93}\text{Zr}_7$ trilayer structure. Time-resolved x-ray magnetic circular dichroism has been developed in transmission, with resonant field excitation at a FMR frequency of 2.3 GHz. Small-angle (to 0.2°), time-domain magnetization precession could be observed directly, and resolved to individual layers through elemental contrast at Ni, Fe, and Co edges. The phase sensitivity allowed direct measurement of relative phase lags in the precessional oscillations of individual elements and layers. A weak ferromagnetic coupling, difficult to ascertain in conventional FMR measurements, is revealed in the phase and amplitude response of individual layers across resonance.

DOI: 10.1103/PhysRevB.74.064409

PACS number(s): 76.50.+g, 76.90.+d, 78.67.Pt, 78.70.Ck

INTRODUCTION

The dynamic properties of soft ferromagnets (FM) play an obviously important role in determining the characteristics of many modern magneto-electronic devices, from giant magneto-resistive (GMR) read heads in hard disk drives to magnetic tunnel junctions and other advanced “spintronic” devices.^{1,2} Precessional dynamics at 1–10 GHz determine the high-speed response of ferromagnetic heterostructures, and present a fundamental limit to increasing data rates in magnetic information storage.^{3,4} FM materials in such structures are often complex, multilayer systems comprised of metallic alloys and other compounds. The interplay between the various layers, or between elemental moments in a single layer, can have a dramatic effect on the high-speed response. It is also the motivation for developing new measurement techniques that can separate the dynamic behavior of these complex structures in a layer-by-layer or element-by-element basis.^{5–13}

Ferromagnetic resonance (FMR) remains a technique of choice for measuring fundamental quantities in precessional dynamics. In FMR, a microwave-frequency rf magnetic field excites motion of the ferromagnetic moments, most strongly where the microwave driving frequency matches the resonant frequency of magnetization precession. The conical precessional motion of \mathbf{M} is indicated schematically in Fig 1(a); for thin films magnetized in plane, dipolar fields compress the orbit into the film plane and the cone becomes strongly elliptical. The response is measured by microwave absorption; the position and width of the resonant absorption provide quantitative information on the resonance and relaxation frequencies, respectively.

The motion of the precession can be parametrized by three variables: the out-of-plane cone amplitude θ_0 , the in-plane cone angle amplitude ϕ_0 , designated here as the cone angle, and the phase of the oscillation ψ . In the case of two weakly coupled systems, a phase lag, $\Delta\psi$, can develop between the two precessing magnetization vectors [Figs. 1(b) and 1(c)]. Sinusoidal motion of the transverse component of \mathbf{M} is expected during precession. As the system is swept

through resonance, by either varying the microwave frequency or modifying the resonant frequency via an external bias field, this motion should go through a maximum in amplitude and a 180° shift in phase. For different, uncoupled FM layers with separate resonances, each responds independently while swept through resonance. For near-identical layers coupled ferromagnetically, resonance frequencies are split into lower-frequency acoustic modes (in-phase for \mathbf{M}_1

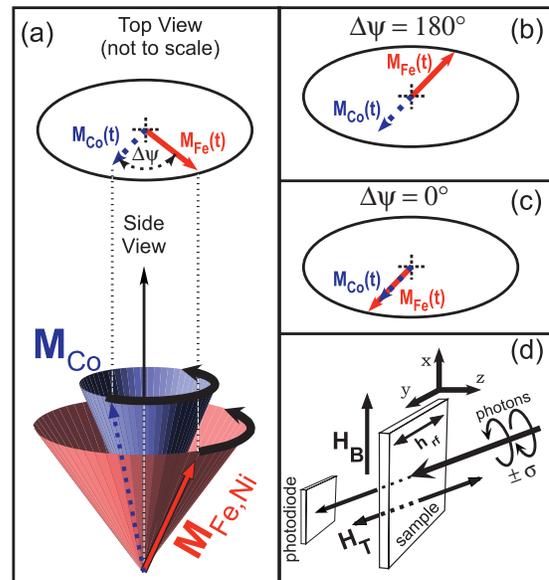


FIG. 1. (Color online) (a) side view of precession cones for $\mathbf{M}_{\text{Fe,Ni}}$ and \mathbf{M}_{Co} showing the different cone angles and top view of phase difference, $\Delta\psi$, between the two vectors. (b) top view indicating out-of-phase difference. (c) top view indicating in-phase condition. (d) Diagram of measurement geometry. The sample is oriented vertically in the x - y plane with the sample normal parallel to z . Photons are incident along the horizontal (in the y - z plane), rotated 38° away from the z axis. Orthogonal Helmholtz coils provide a magnetic field parallel to the film plane along x (bias field \mathbf{H}_B) or parallel to the incident photon direction (transverse field \mathbf{H}_T). Transmitted photons are detected with a photodiode.

and \mathbf{M}_2) and higher-frequency optical modes (out-of-phase for \mathbf{M}_1 and \mathbf{M}_2). Two weakly coupled layers with separated resonances will show mixed behavior.¹⁴

It is worth noting that *layer-resolved* measurements of FMR are particularly relevant to magnetic multilayer structures where the coupling can be varied over a considerable range by judicious selection of nonmagnetic (NM) spacer materials, thickness, and interface roughness. For example, interlayer exchange coupling (IEC) of FM layers through a NM spacer, as in $\text{FM}_1/\text{NM}/\text{FM}_2$, has been investigated since the beginnings of interest in magneto-electronics.¹⁵ Conventional FMR has provided quantitative estimates of parameters such as the magnitude and sign of the ferromagnetic/antiferromagnetic coupling constant A_{ex} between the FM layers through splitting of the coupled resonance frequencies.¹⁴ However, in coupled layers, no measurement technique could directly separate the response of FM_1 from FM_2 ; visible magneto-optical and electronic measurements combine the response of both layers, and separation of the motion is achieved only through models.

In this paper, we demonstrate an element- and time-resolved measurement of ferromagnetic resonance (ETR-FMR) that distinguishes precession in individual layers. The phase and amplitude of the response of individual layers is revealed using time-resolved x-ray magnetic circular dichroism (TR-XMCD), developed in resonant-field pumping and with a transmission geometry. The unique configuration enables orders of magnitude improvement in the resolution of precessional amplitude (to 0.2°) and phase (to 5 ps, or 5° at 2.3 GHz), respectively. Weak ferromagnetic coupling between $\text{Ni}_{81}\text{Fe}_{19}$ and $\text{Co}_{93}\text{Zr}_7$ layers in a $\text{Ni}_{81}\text{Fe}_{19}/\text{Cu}/\text{Co}_{93}\text{Zr}_7$ “pseudo spin valve” trilayer is observed clearly through the amplitude and phase responses of the individual layers, measured separately at the (Ni,Fe) and Co x-ray absorption edges. The weak coupling level (0.01 erg/cm^2 , or $\sim 6 \text{ Oe}$) is not easily detected through complementary microwave absorption measurement.

THEORY

We begin our modeling of a weakly coupled FM trilayer system starting from the Landau-Lifshitz (LL) equation of motion for magnetization dynamics,¹⁶ given in SI as

$$\frac{d\mathbf{M}^{(i)}}{dt} = -\mu_0|\gamma^{(i)}|(\mathbf{M}^{(i)} \times \mathbf{H}_{\text{eff}}^{(i)}) - \frac{\lambda^{(i)}}{|M_s^{(i)}|^2}[\mathbf{M}^{(i)} \times (\mathbf{M}^{(i)} \times \mathbf{H}_{\text{eff}}^{(i)})], \quad (1)$$

where the superscript i indexes FM layer 1 or 2 in the trilayer structure. $\mathbf{M}^{(i)}(t)$ is the magnetization with saturation value $M_s^{(i)}$, $\gamma^{(i)}$ is the gyromagnetic ratio, $\lambda^{(i)}$ is the LL relaxation rate in sec^{-1} and $\mathbf{H}_{\text{eff}}^{(i)}$ is the effective field in the i th FM layer.

In our trilayer system, $\mathbf{H}_{\text{eff}}^{(i)}$ consists of two external fields, \mathbf{H}_B and $\mathbf{h}_{\text{rf}}(t)$, and three internal fields related to the magnetic properties of the films

$$\mathbf{H}_{\text{eff}}^{(i)} = \mathbf{H}_B + \mathbf{h}_{\text{rf}}(t) + \mathbf{H}_D^{(i)} + \mathbf{H}_k^{(i)} + \mathbf{H}_{\text{ex}}^{(i)} \quad (2)$$

where (1) external dc bias field \mathbf{H}_B , (2) external rf microwave field $\mathbf{h}_{\text{rf}}(t)$, (3) demagnetization field $\mathbf{H}_D^{(i)}$, (4) effective magnetocrystalline anisotropy field $\mathbf{H}_k^{(i)}$, and (5) an interlayer coupling field $\mathbf{H}_{\text{ex}}^{(i)}$.

As shown in Fig. 1(d), we take the ferromagnetic trilayer structure to lie in the x - y plane, with z normally directed. The external dc bias field \mathbf{H}_B is taken to be along the x axis, the rf microwave driving field $\mathbf{h}_{\text{rf}}(t)$ is along the y axis, the effective magnetocrystalline anisotropy field $\mathbf{H}_k^{(i)}$ is along the x axis, and the demagnetization field $\mathbf{H}_D^{(i)}$ is along the z axis.

To arrive at an expression for $\mathbf{H}_{\text{ex}}^{(i)}$, we consider the energy per unit area of the coupling between the FM layers, which can be expressed as

$$E_{ex} = -A_{ex} \frac{\mathbf{M}^{(i)} \cdot \mathbf{M}^{(j)}}{M_s^{(i)} M_s^{(j)}}, \quad (3)$$

where the coupling constant A_{ex} has units of energy/unit area and is assumed to be positive for parallel, ferromagnetic coupling between the FM layers and negative for antiparallel, antiferromagnetic coupling between the FM layers. The exchange-coupling field can then be expressed as a derivative of Eq. (3): $\mathbf{H}_{\text{ex}}^{(i)} = -(1/\mu_0 t^{(i)})(\partial E_{ex}/\partial \mathbf{M}^{(i)})$, with the result as

$$\mathbf{H}_{\text{ex}}^{(i)} = \frac{A_{ex}}{\mu_0 M_s^{(i)} t^{(i)}} \cdot \frac{\mathbf{M}^{(j)}}{M_s^{(j)}} = \frac{A_{ex}}{\mu_0 M_s^{(i)} t^{(i)}} \cdot \mathbf{m}^{(j)}, \quad (4)$$

where $t^{(i)}$ is the thickness of the i th FM layer and the superscript j indexes the other FM layer in the trilayer structure. We will consider only a small deviation of the magnetization $\mathbf{M}^{(i)}$ ($i=1,2$) from its equilibrium position $M_s^{(i)} \hat{\mathbf{e}}_{r,i}$

$$\mathbf{M}^{(i)} \approx M_s^{(i)} \hat{\mathbf{e}}_{r,i} + m_{\theta_i} \hat{\mathbf{e}}_{\theta_i} + m_{\phi_i} \hat{\mathbf{e}}_{\phi_i}, \quad (5)$$

where $m_{\theta_i} = M_s^{(i)} \Theta_i$ and $m_{\phi_i} = M_s^{(i)} \sin(\theta_i) \Phi_i$ are small deviations of the magnetizations along the θ_i and ϕ_i directions, respectively. θ_i is the magnetization angle away from the z axis; ϕ_i is the magnetization angle away from the x axis in the x, y plane. Θ_i is expressed in terms of $\Theta_i = \theta_i - 90^\circ$ so that $\Theta_i = 0$ has magnetization in plane. Assuming in-plane magnetization at equilibrium for both FM layers, $\mathbf{m}^{(i)}$ ($i=1,2$) in Eq. (4) can be expressed as

$$\mathbf{m}^{(i)} \approx \hat{\mathbf{e}}_x + \Phi_i \hat{\mathbf{e}}_y - \Theta_i \hat{\mathbf{e}}_z \quad (6)$$

and the demagnetization field $\mathbf{H}_D^{(i)}$ ($i=1,2$) can be written as

$$\mathbf{H}_D^{(i)} \approx M_s^{(i)} \Theta_i \hat{\mathbf{e}}_z. \quad (7)$$

Under the above assumptions, the linearized Landau-Lifshitz equations of motion for small precessional angles (Θ_i and Φ_i for $i=1,2$) can be obtained by expanding Eq. (1) in spherical coordinates and retaining only terms to the first order of Θ_i and Φ_i , shown as the following:

$$\frac{d\Theta_i}{dt} \approx \frac{\lambda^{(i)}}{M_s^{(i)}} H_\theta^{(i)} + \mu_0 |\gamma^{(i)}| H_\phi^{(i)}, \quad (8)$$

$$\frac{d\Phi_i}{dt} \approx -\mu_0|\gamma^{(i)}|H_\theta^{(i)} + \frac{\lambda^{(i)}}{M_s^{(i)}}H_\phi^{(i)}, \quad (9)$$

where $H_\theta^{(i)}$ and $H_\phi^{(i)}$ can be expressed in terms of the Cartesian fields $H_x^{(i)}$, $H_y^{(i)}$, and $H_z^{(i)}$

$$H_\theta^{(i)} \approx -\Theta_i H_x^{(i)} - H_z^{(i)}, \quad (10)$$

$$H_\phi^{(i)} \approx -\Phi_i H_x^{(i)} + H_y^{(i)}. \quad (11)$$

Being more explicit here, we can write the Cartesian fields $H_x^{(i)}$, $H_y^{(i)}$, and $H_z^{(i)}$ as

$$H_x^{(i)} = H_B + H_k^{(i)} + \frac{A_{ex}}{\mu_0 M_s^{(i)} t^{(i)}}, \quad (12)$$

$$H_y^{(i)} = h_{rf}(t) + \frac{A_{ex}}{\mu_0 M_s^{(i)} t^{(i)}} \Phi_i, \quad (13)$$

$$H_z^{(i)} = M_s^{(i)} \Theta_i - \frac{A_{ex}}{\mu_0 M_s^{(i)} t^{(i)}} \Theta_j, \quad (14)$$

where $j=1(2)$ if $i=2(1)$.

Finally, if a harmonic form for the external driving field $h_{rf}(t)$ [*i.e.*, $h_{rf}(t) = h_{rf,0} \exp(i\omega t)$] is assumed, as is the case in force resonance, the linearized Landau-Lifschitz equations of motion for small precessional (Θ_i and Φ_i for $i=1,2$) can be written in matrix form as

$$\frac{d}{dt} \begin{bmatrix} \Theta_1 \\ \Phi_1 \\ \Theta_2 \\ \Phi_2 \end{bmatrix} \approx \mathbf{A} \begin{bmatrix} \Theta_1 \\ \Phi_1 \\ \Theta_2 \\ \Phi_2 \end{bmatrix} + \mathbf{g}(\mathbf{t}), \quad (15)$$

where

$$\mathbf{A} = \begin{bmatrix} -\lambda^{(1)} & -\mu_0|\gamma^{(1)}|\tilde{H}^{(1)} & \frac{\lambda^{(1)}}{M_s^{(1)}}H_{ex}^{(1)} & \mu_0|\gamma^{(1)}|H_{ex}^{(1)} \\ \mu_0|\gamma^{(1)}|(M_s^{(1)} + \tilde{H}^{(1)}) & -\frac{\lambda^{(1)}}{M_s^{(1)}}\tilde{H}^{(1)} & -\mu_0|\gamma^{(1)}|H_{ex}^{(1)} & \frac{\lambda^{(1)}}{M_s^{(1)}}H_{ex}^{(1)} \\ \frac{\lambda^{(2)}}{M_s^{(2)}}H_{ex}^{(2)} & \mu_0|\gamma^{(2)}|H_{ex}^{(2)} & -\lambda^{(2)} & -\mu_0|\gamma^{(2)}|\tilde{H}^{(2)} \\ -\mu_0|\gamma^{(2)}|H_{ex}^{(2)} & \frac{\lambda^{(2)}}{M_s^{(2)}}H_{ex}^{(2)} & \mu_0|\gamma^{(2)}|(M_s^{(2)} + \tilde{H}^{(2)}) & -\frac{\lambda^{(2)}}{M_s^{(2)}}\tilde{H}^{(2)} \end{bmatrix}, \quad (16)$$

$$\mathbf{g}(\mathbf{t}) = \begin{bmatrix} \mu_0|\gamma^{(1)}| \\ \frac{\lambda^{(1)}}{M_s^{(1)}} \\ \mu_0|\gamma^{(2)}| \\ \frac{\lambda^{(2)}}{M_s^{(2)}} \end{bmatrix} h_{rf}(t), \quad (17)$$

and

$$H_{ex}^{(i)} = \frac{A_{ex}}{\mu_0 M_s^{(i)} t^{(i)}}, \quad (18)$$

$$\tilde{H}^{(i)} = H_B + H_k^{(i)} + H_{ex}^{(i)} \ll M_s^{(i)}, \quad (19)$$

$$h_{rf}(t) = h_{rf,0} \exp(i\omega t). \quad (20)$$

EXPERIMENT

The XMCD and ETR-FMR measurements were carried at the circularly polarized soft x-ray beamline 4-ID-C of the Advanced Photon Source (APS) at Argonne National Labo-

raty. Figure 2(a) presents a block diagram of apparatus for *in situ* measurements of conventional FMR and ETR-FMR. The signal from the APS photon bunch clock is first directed through a variable delay and then to a phase-locked frequency synthesizer that upconverts the 88 MHz signal to 2.3 GHz. The amplified output is then directed into a hollow broadband rf resonator to excite the uniform precession of the magnetic thin film sample inside. Two orthogonal sets of Helmholtz coils provide the vertical bias field \mathbf{H}_B or the horizontal transverse field \mathbf{H}_T . Conventional *in situ* FMR measurements are made by varying \mathbf{H}_B and detecting the reflected power in the rf circuit via use of directional coupler and a microwave diode. Figure 2(b) presents a top view of experimental geometry. The sample is rotated 38° with respect to the incident photon direction, which is parallel to the transverse magnetic field \mathbf{H}_T . The transmitted x-ray photons are detected using a standard soft x-ray photodiode.

The trilayer sample, $\text{Ni}_{81}\text{Fe}_{19}(25 \text{ nm})/\text{Cu}(20 \text{ nm})/\text{Co}_{93}\text{Zr}_7(25 \text{ nm})/\text{Cu cap}(5 \text{ nm})$, was grown via UHV magnetron sputtering at a base pressure of 4×10^{-9} Torr. The substrate used was a commercially available Si_3N_4 membrane, 100 nm in thickness. At the transition metal edges of interest, transmission through the substrate is greater than 80%. For ETR-FMR and standard FMR measurements, the

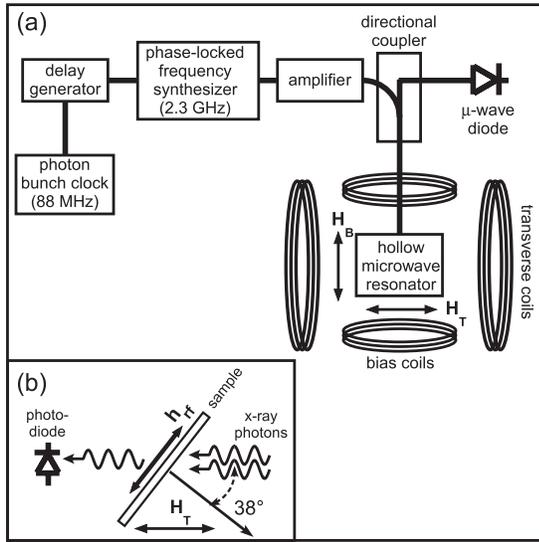


FIG. 2. Block diagram electronics used to generate phase-locked, 2.3 GHz excitation for conventional FMR and ETR-FMR measurements. (b) Top view of experimental geometry.

sample was placed inside the hollow broadband rf resonator.

Static XMCD spectra of Fe, Ni and Co were measured in transmission mode, with strong dichroism signals easily visible on the L_3 and L_2 edges of all FM elements (Fig. 3). Element specific XMCD hysteresis loops, also shown in Fig. 3, were taken as a function of the transverse field \mathbf{H}_T to obtain a calibration for the in-plane rotational magnetization angle ϕ , by tuning the photon energy to the peak XMCD signal (arrows, $h\nu=707.5$ eV for Fe, 851.5 eV for Ni, and 778 eV for Co). The Fe and Ni loops indicate that the $\text{Ni}_{81}\text{Fe}_{19}$ layer switches at a coercive field of 5 Oe; the $\text{Co}_{93}\text{Zr}_7$ layer exhibits a larger coercive field of 7 Oe.¹⁷ All the three XMCD spectra show a negative contribution at the L_3 edge and thus any static coupling between the FM layers is taken to favor parallel alignment of the layers. Also, the thin film sample presents an in-plane anisotropy and the saturation values of each hysteresis loop are taken to be $\pm 90^\circ$.

ETR-FMR timing scans were conducted at a fixed continuous-wave (CW) rf frequency of 2.3 GHz; the CW rf excitation produced a time-varying magnetic field (\mathbf{h}_{rf}) that was in the film plane and transverse to the time-averaged magnetization direction [Fig. 1(d)]. For ETR-FMR, \mathbf{h}_{rf} was phase-locked with the APS photon bunch clock (88 MHz) and thus the instantaneous position of $\mathbf{M}_{\text{Fe,Ni}}$ or \mathbf{M}_{Co} was sampled stroboscopically with the photon pulses once every 26 precession cycles. ETR-FMR timing scans were recorded by tuning the photon energy to the transition metal edge of interest (arrows, Fig. 3), setting the bias field \mathbf{H}_B to the desired point on the conventional *in situ* FMR resonance curve [Fig. 4(a)], and sweeping out the variable delay between the 2.3 GHz CW rf excitation and the APS bunch clock.

RESULTS AND DISCUSSION

An *in situ*, conventional FMR spectrum using the 2.3 GHz, phase-locked CW excitation is presented in Fig

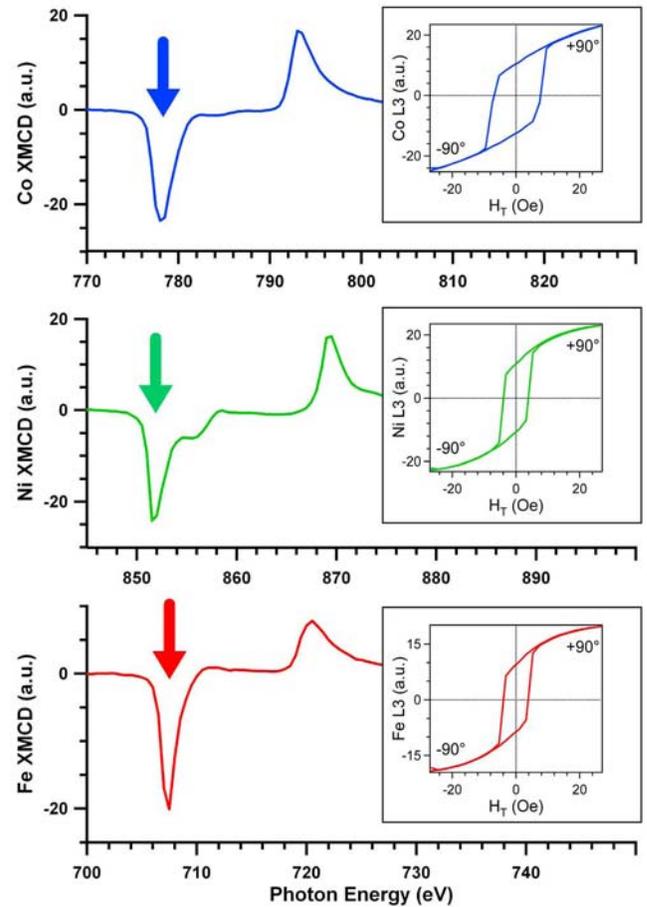


FIG. 3. (Color online) XMCD spectra and element specific hysteresis measurements of the $\text{Ni}_{81}\text{Fe}_{19}/\text{Cu}/\text{Co}_{93}\text{Zr}_7$ /trilayer. The hysteresis curves and ETR-FMR were measured at the photon energies indicated by the arrows (707.5 eV for Fe, 851.5 eV for Ni, and 778 eV for Co). The XMCD signal levels at saturation in the hysteresis curves provide an angular calibration for ETR-FMR measurements.

4(a). A strong resonance, identified by the zero crossing point, is readily apparent at $H_B=40$ Oe. A much weaker inflection is observed near zero bias. In addition, Fig. 4(b) presents measurements of *ex situ*, variable frequency conventional FMR. Plotted in Fig. 4(b) are the resonance field ($H_{B,\text{res}}$) at the square of the variable excitation frequencies (f_p^2). At frequencies higher than the 2.3 GHz used for ETR-FMR measurements, the weak resonance near $H_B=0$ Oe in Fig. 4(a) increases in intensity and is more clearly defined. Two branches are observed in the data in Fig. 4(b), which correspond primarily to separate resonances in the $\text{Ni}_{81}\text{Fe}_{19}$ and $\text{Co}_{93}\text{Zr}_7$ layers. The LL equations for magnetization dynamics can model satisfactorily the data in the absence of coupling between the layers [solid green lines, Fig. 4(b)] or with a very weak FM coupling [dashed magenta lines, Fig. 4(b)].

ETR-FMR timing scans for the $\text{Ni}_{81}\text{Fe}_{19}/\text{Cu}/\text{Co}_{93}\text{Zr}_7$ trilayer were acquired at the field values indicated by the black arrows in Fig. 4(a). Results are exhibited in Fig. 5, where the timing scans have been offset for clarity. All of the data in Fig. 5 exhibit a strong sinusoidal dependence with

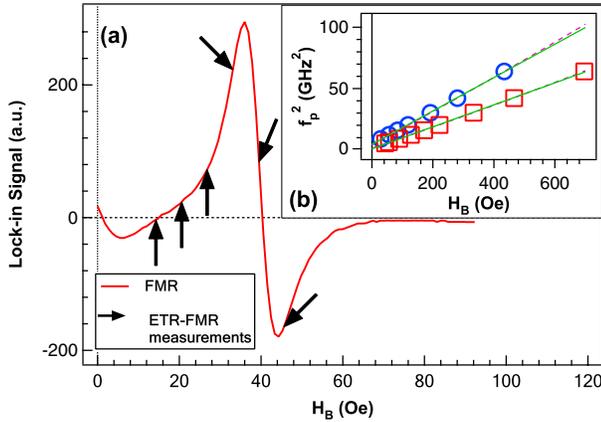


FIG. 4. (Color online) (a) *In situ* FMR measurement at 2.3 GHz. The rf excitation used in the measurement is phase-locked with the synchrotron photon bunch clock at 88 MHz. Note the main resonance indicated by the zero crossing point at $H_B \sim 40$ Oe and a weaker resonance at near zero bias. ETR-FMR measurements were acquired at the field values indicated by the arrows. (b) Position of resonance field $H_{B,res}$, measured with *ex situ*, variable frequency FMR. Two resonances are observed in the trilayer, derived primarily from the $Ni_{81}Fe_{19}$ layer (red squares) and the $Co_{93}Zr_7$ layer (blue circles). See text for description of dashed/solid lines.

time delay and therefore the timing scans in the figure can be fit by simple sinusoidal functions (solid black lines). The amplitudes and phases determined by these fits have a high degree of confidence. Precession amplitudes as small as 0.2° have been measured and the estimated errors are on the order of 0.02° (one standard deviation).¹⁸ The ETR-FMR data also allow for precise determination of the phase of the oscillations; the resolution of the phase determination was 2° – 5° . At 2.3 GHz, this translates to an overall timing resolution of 2–6 ps. The timing resolution achieved in ETR-FMR measurements represents a tenfold to hundredfold improvement over previous TR-XMCD results using pulsed excitations.⁵ Significantly, the phase sensitivity of ETR-FMR enables a

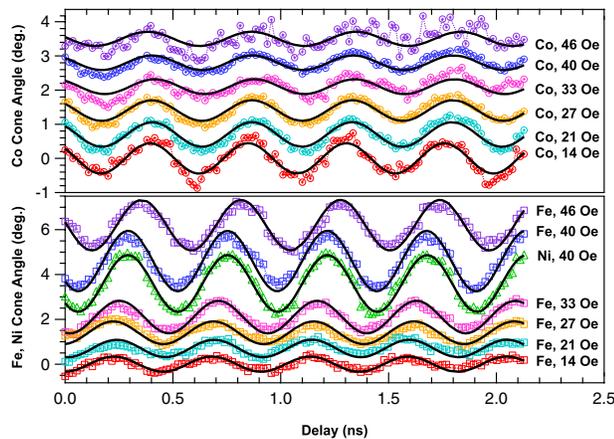


FIG. 5. (Color online) ETR-FMR measurements for Fe, Ni, and Co in the $Ni_{81}Fe_{19}/Cu/Co_{93}Zr_7$ magnetic trilayer. The open symbols are the measured points and the solid black lines are sinusoidal fits to the data. For clarity, the data have been offset vertically.

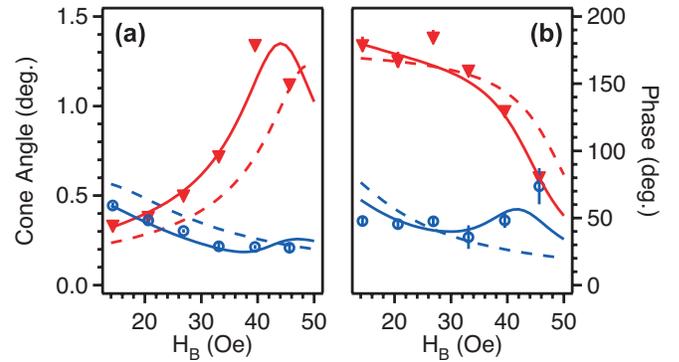


FIG. 6. (Color online) Measured values for the precession cone angle (left panel) and phase of oscillation (right panel) for the Fe (solid red triangles) and Co (open blue circles) moments in the $Ni_{81}Fe_{19}/Cu/Co_{93}Zr_7$ trilayer. The dashed lines are calculated values for the amplitude and phase assuming no coupling ($A_{ex}=0$) between the layers. Solid lines are model calculations that assume weak coupling ($A_{ex}=0.01$ ergs/cm²) between the two FM layers. Estimated errors are indicated by vertical lines; note that some estimated errors are smaller than the size of the symbols.

time resolution an order of magnitude smaller than the intrinsic bunch width of the x-ray photons (~ 60 ps).

In Fig. 5, the top panel presents the time delay scans at different values of H_B for the Co moment oscillation in the $Co_{93}Zr_7$ layer. The bottom panel presents similar data for Fe in the $Ni_{81}Fe_{19}$ film. A representative timing scan for Ni is also presented for $H_B=40$ Oe. The moments of the Fe and Ni in the $Ni_{81}Fe_{19}$ layer are seen to precess in unison within the instrumental resolution of $\sim 0.2^\circ$ in amplitude and ~ 2 ps in phase. The observation of coupled motion between the Fe and Ni moments is a signature of strong exchange coupling within the $Ni_{81}Fe_{19}$ layer that binds the moments of the two FM elements in lockstep, which is consistent with previous TR-XMCD results.⁵

The phase and amplitude parameters extracted from the time delay scans at different bias fields (for $H_B=14, 21, 27, 33, 40,$ and 46 Oe) are presented for Co and Fe in Fig. 6(a) (amplitude) and Fig. 6(b) (phase). In contrast to the identical precession of the Fe and Ni moments within the $Ni_{81}Fe_{19}$ layer, the dynamics *between* FM layers is very different. The clearest trend in Fig. 6(a) is the dramatic increase in amplitude of the Fe oscillations as H_B is increased from 14 Oe (0.34°) to 40 Oe (1.35°), followed by a decline at 46 Oe (1.13°). In contrast, the Co amplitude decreases monotonically. A comparison with Fig. 4(a) indicates that the rapid growth and subsequent decline in the amplitude of the Fe oscillations is a consequence of driving the system through a resonance. Based on the 2.3 GHz FMR data [Fig. 4(a)] and the FMR measurements at higher frequency [Fig. 4(b)], the increase in the Co oscillation amplitude at low values of H_B is consistent with a weak resonance in the $Co_{93}Zr_7$ layer near zero bias.

The changes in the phase of the oscillations in the two layers [Fig. 6(b)] as the system is forced into resonance exhibit a corresponding behavior. From $H_B=14$ Oe through $H_B=46$ Oe the Fe (and Ni) phase undergoes a dramatic

change of $\sim 100^\circ$. This is consistent with the expected phase shift of $\sim 180^\circ$ upon going through a resonance. In contrast, the phase of the Co signal remains largely unaffected. Also, note that the precession of $\mathbf{M}_{\text{Fe,Ni}}$ and \mathbf{M}_{Co} differs in phase by $\sim 120^\circ$ for the lowest value of H_B measured (14 Oe), and that the phase difference decreases to less than 10° at the highest value of H_B measured (46 Oe).

The main features of the changes in the amplitude and phase of precession in $\mathbf{M}_{\text{Fe,Ni}}$ and \mathbf{M}_{Co} are qualitatively consistent with independent resonances in the $\text{Ni}_{81}\text{Fe}_{19}$ and $\text{Co}_{93}\text{Zr}_7$ layers. Closer analysis, however, reveals a weak coupling between the two layers that modifies the resonances of the individual layers. Also shown in Fig. 6 are the calculated values for amplitude and phase of the oscillations based on numerical solutions from the two-particle linearized LL model (coupled linearized LL equations of motion for in $\mathbf{M}_{\text{Fe,Ni}}$ and \mathbf{M}_{Co}). The coupling is parametrized as an effective exchange energy per unit area, A_{ex} in Eq. (4). Results assuming no coupling between the layers (i.e., $A_{ex}=0$, dashed lines) and weak coupling ($A_{ex}=0.01$ ergs/cm², solid lines) are shown in the figures.¹⁹ For the amplitudes of the precession, the assumption of no coupling between the layers overestimates the resonance field $H_{B,res}$ for the main resonance in the $\text{Ni}_{81}\text{Fe}_{19}$ layer which is observed near 40 Oe and also predicts a narrower width for the amplitude than is observed; an assumption of weak coupling more closely matches the observed motion of $\mathbf{M}_{\text{Fe,Ni}}$. For the $\text{Co}_{93}\text{Zr}_7$ layer, weak coupling again provides better agreement with the data as an assumption of uncoupled layers overestimates the amplitude of the precession cone for Co. In the case of the phase of the oscillations, an assumption of independent layers ($A_{ex}=0$, dashed lines) predicts a more gradual decline in the phase of the $\text{Ni}_{81}\text{Fe}_{19}$ layer than is observed, and also does not account for the upturn in the phase of the $\text{Co}_{93}\text{Zr}_7$ layer at high values of H_B . Again, an assumption of weak coupling ($A_{ex}=0.01$ ergs/cm², solid lines) provides better agreement with the measurements. By observing the motion, in both amplitude and phase, of individual layers, ETR-FMR can thus reveal weak coupling much more directly than the conventional, variable frequency FMR presented in Fig. 4(b).

The most likely causes of the weak coupling between $\mathbf{M}_{\text{Fe,Ni}}$ and \mathbf{M}_{Co} are the previously mentioned IEC mechanism or a *Neél* (“orange peel”) type dipolar coupling.^{20,21} *Neél* coupling is more probable as IEC has a considerably shorter interaction range²² and our 20 nm Cu spacer layer is rather thick. Both mechanisms are static interactions that nonetheless can affect the dynamic response of a coupled system. Recently, *dynamic* coupling mechanisms have also been proposed. For example, Tserkovnyak *et al.* propose a

relatively long range, spin pumping mechanism in FM/NM/FM trilayers where precession in one FM layer affects the damping in the other FM layer.^{23,24} Conventional FMR has been used to examine these systems and effects arising from spin-pumping have been detected in resonance line widths.^{25,26} ETR-FMR, however, would be able to measure such effects directly by looking at FM layers individually.

ETR-FMR is inherently a core-level, local-probe technique and as such is well-positioned to illuminate a number of unresolved issues in magnetization dynamics. For example, lagged-response models developed to explain damping mechanisms in magnetization dynamics typically incorporate energy transfer between different reservoirs in a magnetic system; this energy transfer is made manifest via phase lags between different constituents.²⁷ ETR-FMR, which distinguishes among the differences of the magnetization dynamics of individual elements, can examine directly such phase lags in the response of a subsystem. Also, as ETR-FMR is based on the well-developed XMCD technique, *spectroscopic* investigations of dynamics, such as dynamic variations in the orbit-to-spin ratio of magnetic moments or induced magnetism in nonmagnetic layers during precession, are now feasible. Such measurements should provide valuable inputs to emerging first-principles based calculations that seek to move beyond the phenomenological approaches inherent in the LL theory and its extensions.

CONCLUSION

A layer- and time-resolved ferromagnetic resonance, in a “pseudo spin valve” structure of $\text{Ni}_{81}\text{Fe}_{19}/\text{Cu}/\text{Co}_{93}\text{Zr}_7$ has been measured by time-resolved XMCD in transmission mode synchronized with CW microwave excitation at 2.3 GHz. The phase and amplitude of driven FMR precession have been observed magneto-optically, with a very high temporal and rotational sensitivity. More importantly, a weak ferromagnetic coupling between the two FM layers has been revealed much more transparently than any conventional FMR measurements.

ACKNOWLEDGMENTS

The authors thank David J. Keavney (APS) for beamline support. This work was partially supported by the Army Research Office with Grant No. ARO-43986-MS-YIP, and the National Science Foundation with Grant No. NSF-DMR-02-39724. Use of the Advanced Photon Source was supported by the U.S. Department of Energy, Office of Science, Office of Basic Energy Sciences, under Contract No. W-31-109-Eng-38.

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- ¹⁸The maximum cone angle measured was less than 1.4° , which validates the small angle assumption. For uncoupled layers in the small angle regime, the precession amplitude is proportional to the magnitude of the driving rf field.
- ¹⁹Other parameters used in the numerical model are: $\lambda^{(\text{Fe,Ni})} = 1.6$ GHz and $\lambda^{(\text{Co,Zr})} = 5.4$ GHz, $M_s^{(\text{Fe,Ni})} = 780$ kA/m and $M_s^{(\text{Co,Zr})} = 1120$ kA/m, $\mathbf{H}_U^{(\text{Fe,Ni})} = 12$ Oe, and $\mathbf{H}_U^{(\text{Co,Zr})} = 28$ Oe, $g_{\text{eff}}^{(\text{Fe,Ni})} = 2.09$, and $g_{\text{eff}}^{(\text{Co,Zr})} = 2.2$. Similar values were used in the fits for Fig. 4.
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