Concurrent Enhancement of Kerr Rotation and Antiferromagnetic Coupling in Epitaxial Fe/Cu/Fe Structures

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(Received 17 September 1990)

We report the observation of major enhancements in the magneto-optical polar Kerr rotation in the Fe/Cu/Fe system. These enhancements are as much as a factor of 2 greater than those anticipated due to recently reported plasma-resonance effects. A close connection between these enhancements and antiferromagnetic coupling between the two Fe films is established by the observation of concurrent oscillations in the magnitude of these two effects as a function of Cu thickness.

PACS numbers: 75.50.Ee, 75.50.Rr, 75.70.Cn, 78.20.Ls

The search for ways to enhance the magneto-optical Kerr rotation in thin films has become a major focus of research in recent years because of the great impact such enhancements could have on the development of improved magneto-optical recording media.[1-9] Among the most important recent developments in this area is the report that in polycrystalline Fe/Cu films the coupling between the incident photon and the plasma edge of Cu can give as much as a factor-of-2 enhancement in Kerr rotation as compared to pure Fe.[6] In the present work, we have investigated the saturation Kerr rotation in epitaxial Fe/Cu/Fe structures and have found that in addition to the plasma-edge effect there is a further enhancement of the Kerr rotation, by as much as a factor of 2. This further enhancement in the saturation Kerr rotation is largest at just the Cu-spacer-layer thicknesses for which the antiferromagnetic exchange coupling between the two Fe films is largest. The strength of the antiferromagnetic interlayer exchange coupling oscillates as a function of Cu-spacer-layer thickness with a spatial period of 13.5 Å. The Kerr rotation in the magnetically saturated state also oscillates as a function of Cu-spacer-layer thickness with a period of 13.5 Å. This novel effect suggests an oscillatory character of the electronic structure in the saturated state as a function of Cu-spacer-layer thickness.

Antiferromagnetic coupling through nonmagnetic spacer layers between individually ferromagnetic segments has been a topic of much interest recently.[10-21] Investigations of the zero-field magnetic structure in Fe/Cr multilayers revealed in-plane antiferromagnetic alignment of Fe layers on either side of Cr spacer layers.[10-13] The application of a sufficiently large in-plane magnetic field to single-crystal multilayered structures of Fe/Cr induced ferromagnetic alignment of the Fe layers and revealed unexpectedly high values of saturation magnetoresistance.[12,13] More recently, oscillations in the strength of the interlayer magnetic exchange coupling with spacer-layer thickness have been observed in Fe/Cr, Co/Ru, Co/Cr, and Fe/Cu multilayers.[14,21] Moreover, the authors discovered oscillations in the magnitude of the saturation magnetoresistance of these superlattices that were commensurate with the oscillations in the strength of antiferromagnetic coupling.[14] In this Letter, we report a new effect commensurate with oscillations in the strength of antiferromagnetic coupling: oscillations in an enhancement of the Kerr rotation.

To investigate the coupling between two ferromagnetic 3-ML fcc-Fe films (ML denotes monolayer) separated by a Cu spacer, we grew sandwich structures of the form $Cu_{60}Fe_3Cu_xFe_3Cu(100)$. Spin-polarized neutron reflection studies of single ultrathin fcc-Fe films sandwiched between Cu(100) demonstrate a large ferromagnetic dipole moment per atom over only a small range of Fe thicknesses.[22] In particular, 3-ML-thick fcc-Fe films grown epitaxially on a Cu(100) substrate with a 60-ML Cu(100) protecting overlayer exhibit an average magnetic dipole moment per atom close to that of bulk bcc Fe.[22] The growth procedure for the films studied here began by depositing ~10 ML Cu on a clean well-annealed, single-crystal Cu(100) surface at 450 K. This homoepitaxy buries residual imperfections, thereby improving surface quality, and increasing the perpendicular magnetic anisotropy of the final structures.[23] Atomically sharp Fe/Cu interfaces were achieved by employing the growth technique of Steigerwald and co-workers which circumvents the tendency of Fe to agglomerate and Cu to segregate during room-temperature deposition of Fe on Cu(100).[24]

Room-temperature polar Kerr rotation (θ_K) and ellipticity (ε_K) hysteresis loops were recorded *ex situ* for each of our samples at 633 nm using a polarization modulation technique.[25,26] He-Ne laser light, passed through a photoelastic modulator (PEM) and a hole in the pole of an electromagnet, strikes a sample at 1° to the sample normal and returns through the hole. The modulation axis of the PEM is in the plane of incidence and the light incident on the PEM is linearly polarized at 45° to the plane of incidence. A prism refracts the specularly reflected light (in the same plane of incidence) onto a beam-splitting Thompson polarizer. Light polarized in the plane of incidence is directed onto a photodiode. The incident polarization state and the analyzer angle are chosen to optimize both θ_K and ε_K signals, which

are given by $\varepsilon_K = -[2\sqrt{2}J_1(A)]^{-1}S_f/S_{dc}$ and $\theta_K = [2\sqrt{2}J_2(A)]^{-1}S_{2f}/S_{dc}$, where $J_n(A)$ is the nth-order Bessel function evaluated at a modulator phase amplitude such that $J_0(A)=0$, S_{dc} is the light level on the detector, and S_f and S_{2f} are the fundamental and second-harmonic signals with $f=50$ kHz the modulator frequency. S_f and S_{2f} are measured in differential mode using two lock-in amplifiers. A quarter-wave plate is included before the analyzer for calibration of the ellipticity and Kerr rotation.[27]

Ellipticity and Kerr rotation hysteresis data are shown in Fig. 1 for a 15-ML Cu spacer thickness, i.e., $Fe_3Cu_{15}Fe_3$ grown on Cu(100), and coated by 60 ML Cu. By convention, the intrinsic saturation ε_K and θ_K are defined with the magnetic field directed into the film, along the direction of the incident light (positive magnetic field in Fig. 1). For all Cu spacer thicknesses studied here (1–30 ML) the measured saturation ellipticity and saturation Kerr rotation are positive quantities.

The magneto-optical polar Kerr rotation in Fe/Cu has been successfully modeled[6] by numerical calculation using the bulk optical constants of Fe and Cu for left and right circularly polarized light.[28,29] Straightforward extension of this model permits calculation of both saturation ε_K and θ_K in our $Cu_{60}Fe_3Cu_xFe_3Cu(100)$ structures. In the absence of measured off-diagonal dielectric tensor elements for fcc Fe, we calculated the saturation ε_K and θ_K for our samples using values of the tensor elements available at 633 nm for bcc Fe.[28] In Fig. 2 the results of this calculation (the dashed line) may be compared with the measured magneto-optical activity (data points) expressed as the maximum saturation Kerr rotation $(\varepsilon_K^2+\theta_K^2)^{1/2}$. The maximum Kerr rotation can be directly measured by adjustment of a phase plate, placed between the PEM and the sample, with the retardation axis in the plane of incidence.

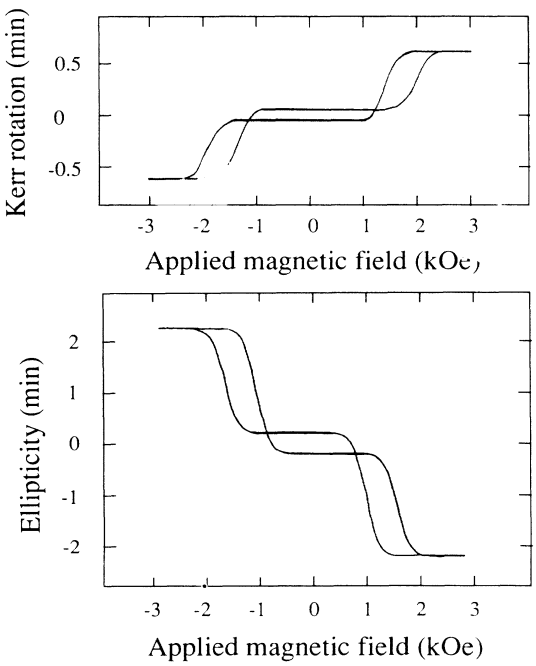

FIG. 1. The polar Kerr rotation and ellipticity hysteresis loops taken at 633 nm for epitaxial fcc-$Fe_3Cu_{15}Fe_3$ grown on a Cu(100) substrate and protected by 60 ML Cu.

In Fig. 2, the data exhibit a magneto-optical activity enhanced by as much as a factor of 2 over that predicted by the calculation using the optical constants of bulk Fe and Cu (which describe the recently reported plasma-edge enhancements[6]). This additional enhancement could be due to differences in the off-diagonal dielectric tensor elements between fcc Fe and bcc Fe, and/or to changes in the dielectric properties of the fcc Fe induced by coupling between the Fe layers in our $Cu_{60}Fe_3Cu_x$-$Fe_3Cu(100)$ structures. To put the size of this additional enhancement in perspective it may help to note that we achieve a maximum Kerr rotation of 4 min (the maximum in Fig. 2) with a total of only 3 ML+3 ML=6 ML of Fe, while the recently reported enhancement of Kerr rotation in the Fe/Cu system implies that approximately 30 ML Fe are required at 633 nm (Ref. 6) (12 ML if the ellipticity is also taken into account[30]). In the Fe/Au(100) system a maximum Kerr rotation of 2.2 min was recently found at ~70 ML Fe.[31]

The peak in the maximum saturation Kerr rotation in the range of 6–8 ML Cu in Fig. 2 coincides with dramatic changes in the shape of the hysteresis loops. This effect is illustrated in Fig. 3, where only the ellipticity loops are displayed since the Kerr loops have the same general shape (as shown in Fig. 1). The shape of the $Fe_3Cu_6Fe_3$ and $Fe_3Cu_8Fe_3$ loops are consistent with antiferromagnetic coupling of the two 3-ML fcc-Fe films. Some insight into the importance of exchange coupling for the peak at 6–8 ML Cu in Fig. 2 may be gained by comparing this peak to twice the signal obtained for a single 3-ML fcc-Fe film in the $Cu_{60}Fe_3Cu(100)$ structure (see the arrow shown in Fig. 2).

To interpret our results in the simplest way we first

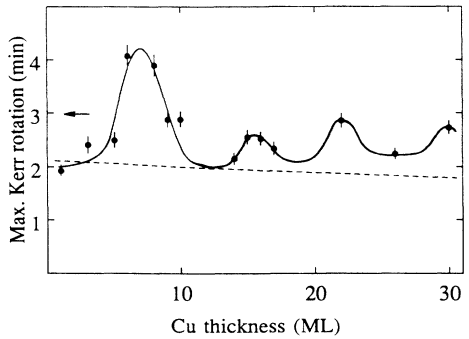

FIG. 2. The maximum Kerr rotation $[(\varepsilon_K^2+\theta_K^2)^{1/2}]$ vs the Cu-spacer-layer thickness (x) in epitaxial fcc-$Cu_{60}Fe_3Cu_x$-$Fe_3Cu(100)$ structures. The solid curve is a guide to the eye. The dashed line is the behavior predicted by a model calculation for our layered structures using the bulk optical constants of Cu and Fe. The arrow illustrates twice the signal from a $Cu_{60}Fe_3Cu(100)$ structure.

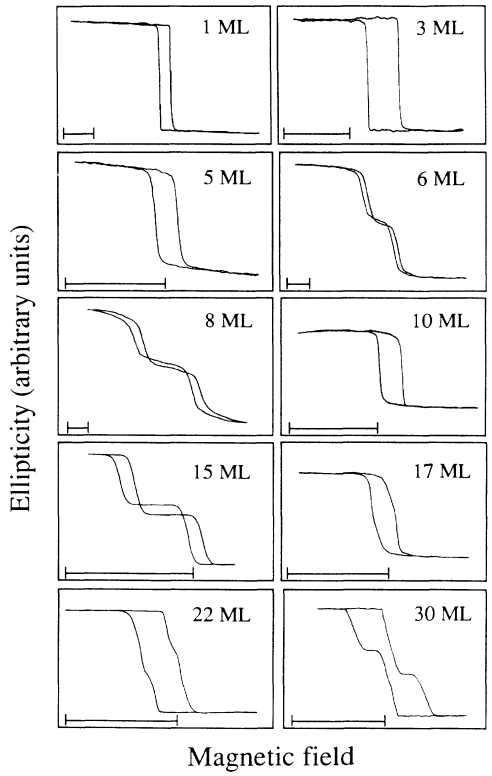

FIG. 3. The ellipticity hysteresis loops for epitaxial fcc-$Cu_{60}Fe_3Cu_xFe_3Cu(100)$ structures. The thickness of the Cu spacer layer, x, is given in the upper-right-hand corner of each box. Different magnetic-field scales were used to best display the loop shapes. The line segment in each box represents 5 kOe (0.5 kOe in the bottom-right box).

write the total system energy in the saturated perpendicular state as $2J-2M_sHd$, where J (>0) is the interlayer antiferromagnetic exchange coupling per unit area, M_s is the saturation magnetization (assumed identical for both layers), d is the thickness of each Fe layer, and H is the magnetic field applied perpendicular to the sample plane. In the antiferromagnetic perpendicular state the total energy in the applied magnetic field is $-2J$. Therefore, the system will make a transition to the antiferromagnetic state at an applied magnetic field given by $H_{AF}=2J/M_sd$, provided a zero-coercivity pathway exists between these states. For nonzero-coercivity transitions, H_{AF} is identified as the field at the loop center, as shown in Fig. 1. If this transition proceeds by coherent rotation of the magnetic moments, it will be necessary to modify the relation between H_{AF} and the magnitude of the interlayer exchange coupling.

Using our simple model, we illustrate the dependence of exchange coupling on Cu-spacer-layer thickness by plotting H_{AF} as shown in Fig. 4. Referring to this figure, there is an oscillation in the magnitude of the interlayer exchange coupling with a period D of 7.5 ± 0.5 ML Cu. Observation of antiferromagnetic coupling at expected peaks positions, i.e., at $3D$ (22.5 ML) and $4D$ (30 ML),

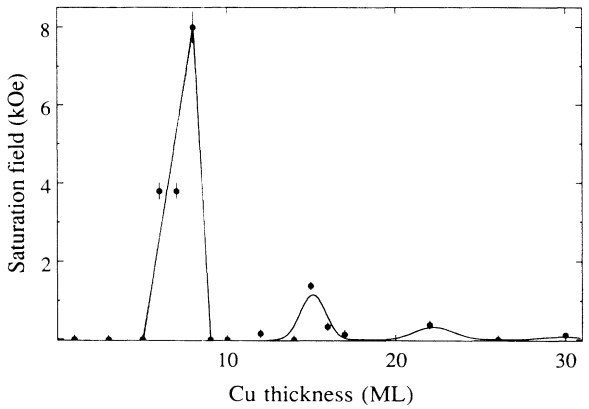

FIG. 4. The saturation field in epitaxial fcc-$Cu_{60}Fe_3Cu_xFe_3Cu(100)$ structures vs the Cu-spacer-layer thickness x. The solid curve is a guide to the eye.

demonstrates that the range of this magnetic exchange coupling is at least 30 ML Cu. The phase of this oscillation would situate an antiferromagnetic maximum "centered" at approximately zero Cu-spacer-layer thickness. It may be significant that we observe no polar Kerr rotation or ellipticity for a 6-ML Fe film, i.e., $Cu_{60}Fe_6Cu(100)$, as expected if antiferromagnetic coupling occurs and exceeds our available field strength (23 kOe).

Using $J=M_sH_sd/2$, and a measured value of $(1.9\pm0.3)\mu_B$ per atom in our earlier fcc-$Cu_{60}Fe_3Cu(100)$ structures,[22] we calculate a maximum exchange-coupling energy (for the 8-kOe saturation field shown in Fig. 4) of 0.32 ± 0.05 erg/cm^2. Unfortunately, if the coupling becomes ferromagnetic, this technique will not provide any information on the size of J. However, we have preliminary evidence which might suggest that the oscillation in exchange coupling is not only in the strength of the antiferromagnetic coupling, but also involves alternation of the sign of the coupling. By measuring the initial magnetization curves immediately after growing these samples, we established that the as-deposited magnetic structure is antiferromagnetic when J is positive and is ferromagnetic, in some cases, when the hysteresis loops have a simple square appearance (as shown in Fig. 3, for a Cu spacer thickness of 3 ML).

Comparing Figs. 2 and 4, there are clear peaks in both H_{AF} and the saturation magneto-optical activity centered around 7.5 ML Cu. There is also evidence of a small peak in the saturation magneto-optical activity centered at 15 ML in Fig. 2 which matches a corresponding peak in H_{AF} at the same Cu-spacer-layer thickness, as shown in Fig. 4. We also note that the antiferromagnetically coupled samples at $3D$ and $4D$ Cu-spacer-layer thickness exhibit relatively high values of maximum Kerr rotation in Fig. 2 (high relative to the downward trend predicted by the model calculation, viz., the dashed line, and the signal from the sample at $3.5D$ Cu-spacer-layer thickness). These concurrent oscillations in magnetic and optical properties suggest a gen-

eral sensitivity of electronic structure to Cu-spacer-layer thickness. A possible source of similar oscillations seen in the interlayer magnetic coupling of Gd/Y [32] and Co/Ru [14] has been proposed to be the Ruderman-Kittel-Kasuya-Yosida (RKKY) indirect exchange, with the reservation in the case of Co/Ru [14] (as in our case) that the phase of the oscillation in the exchange coupling favors antiferromagnetic alignment in the limit of zero spacer-layer thickness, and that the period is longer than expected for an RKKY interlayer coupling. This indirect exchange coupling might modify the distribution of available magneto-optically active transitions and thus provide a mechanism for the observed enhancements in maximum Kerr rotation.

In summary, we have investigated the magnetic coupling between two 3-ML-thick ferromagnetic fcc-Fe films separated by a Cu spacer layer in epitaxial Cu_{60}-$Fe_3Cu_xFe_3Cu(100)$ structures specially prepared to have atomically sharp interfaces. We have observed concurrent oscillations in the interlayer exchange-coupling strength and the zero-ellipticity polar Kerr rotation. The oscillation in exchange coupling covers at least four periods (x =0–30 ML), and has a phase consistent with antiferromagnetic coupling in the limit of zero Cu-spacer-layer thickness. Preliminary evidence indicates that the oscillation in exchange coupling is not only in magnitude, but may involve alternation between antiferromagnetic and ferromagnetic states. Observation that the strength of interlayer exchange coupling correlates with the magnitude of magnetoresistance in Co/Ru, Co/Cr, and Fe/Cr superlattices, [14] and an optical property in epitaxial fcc-$Cu_{60}Fe_3Cu_xFe_3Cu(100)$ structures (this work), suggests a general sensitivity of electronic properties in these layered magnetic systems to long-range coupling effects. Also, we note the similarity between two distinct magnetic/nonmagnetic layered systems: Co/Ru [14] and our Fe/Cu/Fe, both of which exhibit oscillations in the magnitude of the exchange coupling with periods of ~7 ML, and have phases consistent with antiferromagnetic coupling in the limit of zero spacer-layer thickness. Finally, our samples exhibit a major enhancement in the Kerr rotation over that expected on the basis of plasma-resonant absorption. Close correlation of this enhancement with the strength of the interlayer magnetic coupling suggests a common mechanism for these effects.

The authors would like to thank J. Cochran and B. Heinrich for helpful discussions.

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