Short-Period Oscillations in the Interlayer Magnetic Coupling of Wedged Fe(100)/Mo(100)/Fe(100) Grown on Mo(100) by Molecular-Beam Epitaxy

Z. Q. Qiu, J. Pearson, A. Berger,[a] and S. D. Bader
Material Science Division, Argonne National Laboratory, Argonne, Illinois 60439

(Received 23 December 1991)

(100)-oriented Fe/Mo/Fe sandwiches were grown by molecular-beam epitaxy onto a Mo(100) crystal and were investigated *in situ* by means of the magneto-optic Kerr effect. The Fe layers are each 14 monolayers (ML) thick, and the intervening Mo is wedge shaped to facilitate the study of the magnetic coupling between the two Fe films as a function of Mo thickness in the range 1-18 ML. Five oscillations between antiferromagnetic and ferromagnetic coupling were observed with a periodicity of ~3 ML of Mo.

PACS numbers: 75.50.Rr, 75.70.Ak, 75.70.Cn

One of the most exciting recent findings in the field of magnetic thin films was the discovery of the antiferromagnetic (AF) coupling between two Fe films across intervening Cr layers [1]. Subsequent studies revealed that the AF coupling persists in Fe/Cr superlattices and results in a giant magnetoresistance which could be valuable in numerous applications [2]. Much effort has been made to search for other systems which show AF coupling behavior, and many have been found that belong to the same class of materials as Fe/Cr. Examples include Fe/Cu [3], Fe/Mo [4], Co/Ru [5], Co/Cr [5], and Co/Cu [6], etc. These systems exhibit oscillatory behavior between AF and ferromagnetic coupling, with the oscillation periodicity at a value of ~10-15-Å thickness of the nonferromagnetic component. While it is most likely that the Ruderman-Kittel-Kasuya-Yosida- (RKKY-) type interaction is responsible for the oscillatory coupling [7], it is still not quite clear why the periodicity is rather long and similar for all of the above systems, although roughness is believed to play an important role. The value of the oscillation period is greater than the typical RKKY periodicity of ~1-3 monolayers (ML). Thus, it becomes important to search for the existence of short-period oscillations in these systems. Two systems, Fe/Cr [8,9] and Fe/Mn [10], were first found to exhibit short-period (~2 ML) oscillations. The samples of both of those systems were fabricated by evaporation onto flat Fe whiskers, and the nonferromagnetic spacer layer was grown using a moving mask to form a wedge. The probe beam then measures the magnetic coupling for different thicknesses of the spacer layer as it scans across the sample. The 2-ML periodicity was attributed to the direct *d-d* exchange interaction between Fe and Cr [9] (Mn) [10] and the AF nature of the Cr (Mn) spacer layer, rather than to the RKKY interaction between the *d* electrons of Fe and the *s* electrons of Cr or Mn. However, 2-ML short-period oscillations were recently reported on an Fe/Au system [11] and the spacer layer Au is not an antiferromagnetic element like Cr and Mn. Obviously more studies are needed in order to better understand the underlying mechanisms for the magnetic coupling and the relationship between the long- and short-period oscillations. In this Letter we report the results of a study on a new wedged system: Fe(100)/Mo(100)/Fe(100) sandwiches grown on Mo(100). We find that this system also shows short-period oscillations between AF and ferromagnetic coupling with a periodicity of ~3 ML of Mo.

The Fe/Mo/Fe sandwiches were grown by molecular-beam epitaxy (MBE) onto a Mo(100) single-crystal substrate at room temperature in an ultrahigh vacuum (UHV) chamber of base pressure 2×10^{-10} Torr. The chamber is equipped with low-energy electron diffraction (LEED), Auger electron spectroscopy, an Ar-ion sputtering gun for substrate cleaning, and a UHV-compatible superconducting magnet. The Mo(100) substrate is a single-crystal disk of ~2 mm thickness and ~10 mm diameter. The substrate was mechanically polished to a ~1-μm diamond-paste finish and ultrasonically cleaned in methanol before its introduction into the UHV chamber. Then cycles of 1-keV Ar^+ sputtering and annealing at 700 °C for ~30 min were used to improve the surface quality. Auger spectroscopy was used to confirm the cleanliness of the surface. LEED indicated that a well-defined Mo(100) surface was formed [Fig. 1(a)].

The Fe and Mo layers were grown at room temperature by evaporating an Fe wire from an alumina crucible surrounded by W heating wire [12], and by evaporating a 2-mm-diam Mo wire held at 4 kV from an electron gun whose design is based on that of Jonker [13]. The typical

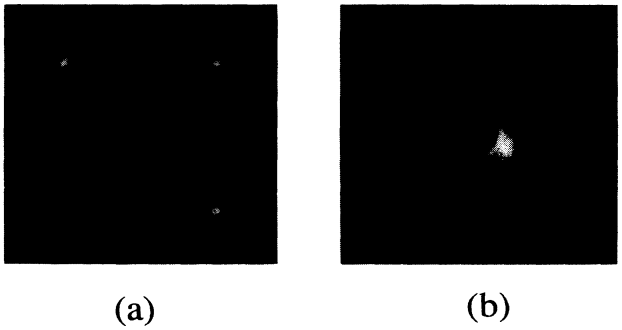


FIG. 1. The LEED patterns for (a) the Mo(100) surface, and (b) a Fe(14 ML)/Mo(*n* ML)/Fe(14 ML) sandwich.

evaporation rate for both Fe and Mo, as monitored using a quartz-crystal oscillator, was ~1-2 Å/min, and the pressure during growth remained $<3\times10^{-10}$ Torr. A 14-ML Fe film was first grown onto the Mo(100) substrate, followed by the growth of a Mo wedge. The wedge was created by placing a Ta mask above and in front of the specimen to block part of the evaporated Mo beam, and by slowly translating the specimen along the [010] crystal direction behind the mask. The speed of the specimen motion during growth was typically ~0.5-1.0 mm/min so that with an evaporation rate of 1-2 Å/min, the Mo wedge would have a thickness gradient of ~1-2 Å/min along the [010] direction. After the growth of the Mo wedge, another 14-ML Fe film would be deposited to complete the Fe (14 ML)/Mo(n ML)/Fe(14 ML) sandwich structure. Subsequent annealing of the sample at 150 °C for ~30 min improved the surface smoothness. Four samples were fabricated in this manner to cover the Mo thickness range of 1-18 ML. The LEED pattern of Fig. 1(b) shows that the Fe/Mo/Fe sandwich forms as a (100)-oriented single crystal.

The magnetic coupling at room temperature between the two Fe films across the Mo spacer layer was studied *in situ* by means of the surface magneto-optic Kerr-effect (SMOKE) technique. The incident beam from a He-Ne laser was focused onto the sample surface (Fig. 2) by an optical lens and polarized in the plane of incidence (p polarization). After reflection from the specimen, the beam was focused again by another optical lens onto a photodiode detector. A quarter-wave plate was used to remove the birefringence of the UHV window, and an analyzing polarizer was set at $\delta\sim1°$ from extinction. The external magnetic field H was applied in the film plane and in the plane of incidence (longitudinal Kerr effect), and along the [001] direction of the sample. The light intensity detected by the photodiode was recorded as a function of the applied magnetic field to generate magnetic hysteresis loops of height ΔI. As discussed in an earlier paper [14], the quarter-wave plate introduces an additional 90°-phase shift between the s and p components of the reflected light so that the quantity measured is the Kerr ellipticity ϕ'':

$$\phi''=(\delta/4)\Delta I/I_0\,,$$

where I_0 is the light intensity at zero magnetization.

Typical hysteresis loops of the Fe(14 ML)/Mo(n ML)/Fe(14 ML) sandwiches for ferromagnetic- and AF-coupled cases are shown in Figs. 3(a) and 3(b). The corresponding MO thicknesses are 6.3 and 7.6 ML, respectively. It should be mentioned that the magnetization is in the plane of the film and no signal perpendicular to the film plane (polar Kerr effect) was observed in the same magnetic-field range. While the hysteresis loop characteristic of ferromagnetism [Fig. 3(a)] is square with low saturation field and high remanent magnetization, the loop characteristic of AF coupling [Fig. 3(b)] shows a plateau in low field with negligible remanence and a rather abrupt switching at high field. This AF-coupling character is quite different from that of sputtered (110)-textured Fe/Mo superlattices, which show a continuous rotation of magnetization towards saturation in AF-coupled samples [4]. The plateau in Fig. 3(b) is due to the AF coupling which aligns the magnetic moments of the two Fe films in an antiparallel fashion to give a zero net magnetization. The near-complete cancellation of the Kerr signal at low field indicates that the thicknesses of the two Fe films are virtually identical. At high field, the external magnetic field overcomes the AF coupling and aligns the magnetic moments of the two Fe films parallel, yielding a net magnetization characteristic of ferromagnetic saturation. An interesting feature appears in the transition region from the antiparallel-to-

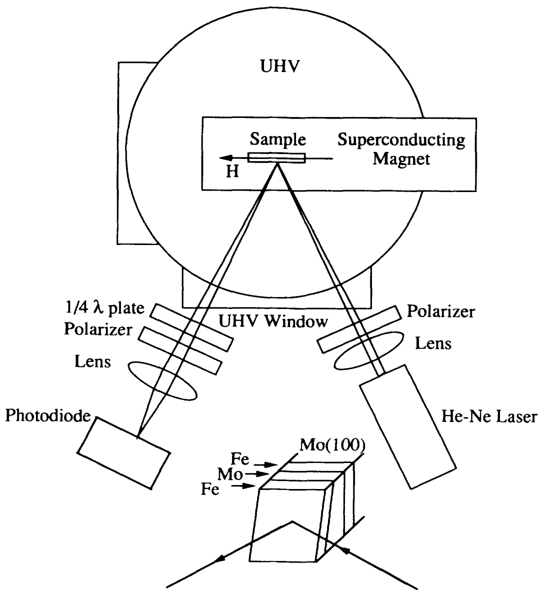


FIG. 2. Schematic drawing of the experimental setup.

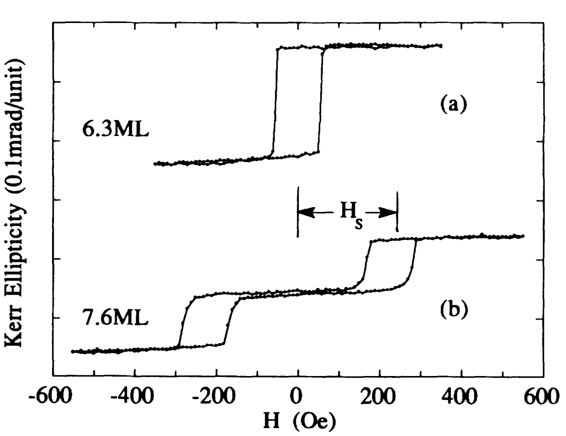


FIG. 3. Typical (a) ferromagnetic- and (b) AF-coupled hysteresis loops for an Fe(14 ML)/Mo(n ML)/Fe(14 ML) sandwich with (a) $n=6.3$ ML and (b) $n=7.6$ ML.

parallel alignment of the magnetization in Fig. 3(b). The rather sharp transition and the zero magnetization at the plateau region is characteristic of a parallel-to-antiparallel switching process between the magnetic moments of the two Fe layers, as has been reported for the Fe/Cu(100) system whose easy axis is perpendicular to the film plane [3]. But for thicker Mo films (Fig. 4), the plateau in the AF-coupling hysteresis loop no longer gives zero net magnetization, suggesting that 90° switching between the magnetic moments in the two Fe layers starts to occur, as has been observed in the Fe/Au system and attributed to a quadratic term in the interlayer coupling [11]. The field at which the switching occurs is roughly the offset of the hysteresis loop from zero field (denoted as H_S and shown in Fig. 3). For the ferromagnetic case the offset is simply zero. The magnitude of the Kerr signal decreases smoothly as the thickness of Mo increases, as calculated using the formalism of Zak *et al.* [15]. There is no evidence for a magneto-optical enhancement in AF-coupled samples, such as has been reported in Ref. [3] for Fe/Cu(100) sandwiches.

In order to investigate the oscillatory behavior, we scanned the laser (with the beam size < 0.2 mm) along the length of the wedge and recorded the hysteresis loops at different positions. Five oscillations of the magnetic coupling were observed in the 1-18 ML range of Mo thickness. Representative hysteresis loops from each oscillation are plotted in Fig. 4, and the switching field is plotted in Fig. 5. We estimate the AF coupling strength in a similar way as in Ref. [9], such that the switching field (H_S) just overcomes the AF coupling between the two Fe films, i.e.,

$$J \approx M_S H_S d \,,$$

where J is the coupling strength per unit area between the two Fe films, M_S is the saturation magnetization, and d is the thickness of the Fe layer. Using this expression, we estimate the coupling strength at $d_{\mathrm{Mo}} \approx 5$ ML to be ~ 0.2 mJ/m^2, which is about 25% of the coupling strength in the Fe/Cr system [9].

From Fig. 4 we see that the oscillation is not only in the coupling strength but it is also between the AF coupling and the ferromagnetic (or zero) coupling. We are not able to distinguish the ferromagnetic coupling and the possibility of noncoupling just from the shape of the hysteresis loop. This result is different from the SMOKE studies on Fe/Cr [9] and Fe/Mn [10] wedged samples, which always showed AF coupling with only the coupling strength oscillating.

Figure 5 shows that the oscillation periodicity is ~ 3 ML of Mo, suggesting that the magnetic coupling is due to the RKKY interaction rather than to a direct exchange interaction between magnetic moments on Fe and Mo sites. The latter would give 2-ML periodicity and would require the extreme assumption that the Mo possess moments and be an antiferromagnet. The 3 ML (4.7 Å) periodicity is much shorter than the periodicity of ~ 11 Å in sputtered Fe/Mo superlattices [4], and agrees with the prediction of Levy, Fry, and Ethridge who recently calculated the periodicity for the Fe/Mo(100) system by assuming an RKKY exchange coupling with slight interface roughness [16]. Another interesting feature is that the first AF peak that we observe appears at ~ 5 ML of Mo. Based on the ~ 3-ML oscillation periodicity, there should be another AF coupling peak at ~ 2 ML of Mo, but it was not observed. This could be due to the interfacial roughness between Fe and Mo, since even with 1-ML roughness it is easy to produce pinholes in a 2-ML Mo layer. Pinholes in the Mo layer would allow the two Fe

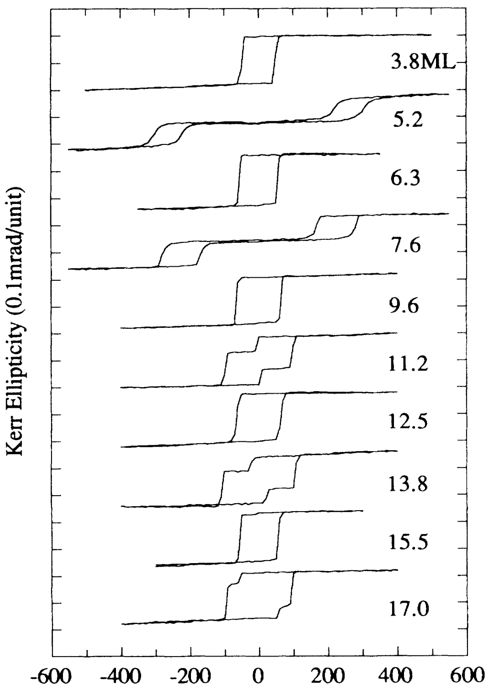


FIG. 4. Hysteresis loops for different thicknesses of Mo, as denoted on the right.

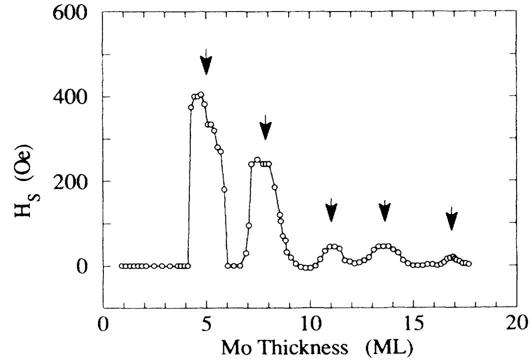


FIG. 5. The switching field, as defined in Fig. 3, vs Mo thickness.

films to join together into a single ferromagnetic film.

In summary, Fe/Mo/Fe(100) sandwiches with the Mo spacer layer fabricated to be wedge shaped have been grown by MBE and investigated by the SMOKE technique. We found that the exchange coupling of the two Fe films separated by Mo oscillates between ferromagnetism and AF coupling as a function of the Mo thickness. Five oscillations were observed at room temperature with the oscillation periodicity of ∼3 ML of Mo.

The work was supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under Contract No. W-31-109-ENG-38.

---

[a] Permanent address: Institute für Grenzflächenforschung und Vakuumphysik der Kernforschungsanlage, Jülich, Germany.

[1] P. Grünberg, R. Schreiber, Y. Pang, M. B. Brodsky, and H. Sowers, Phys. Rev. Lett. **57**, 2442 (1986).

[2] M. N. Baibich, J. M. Broto, A. Fert, F. Nguyen Van Dau, F. Petroff, P. Etienne, G. Creuzet, A. Friederich, and J. Chazelas, Phys. Rev. Lett. **61**, 2472 (1988).

[3] W. R. Bennett, W. Schwarzacher, and W. F. Egelhoff, Jr., Phys. Rev. Lett. **65**, 3169 (1990).

[4] M. E. Brubaker, J. E. Mattson, C. H. Sowers, and S. D. Bader, Appl. Phys. Lett. **58**, 2306 (1991).

[5] S. S. P. Parkin, N. More, and K. P. Roche, Phys. Rev. Lett. **64**, 2304 (1990).

[6] D. Pescia, D. Kerrkmann, F. Schumann, and W. Gudat, Z. Phys. B **78**, 475 (1990); S. S. P. Parkin, R. Bhadra, and K. P. Roche, Phys. Rev. Lett. **66**, 2152 (1991).

[7] Y. Wang, P. M. Levy, and J. L. Fry, Phys. Rev. Lett. **65**, 2732 (1990); D. M. Edwards, J. Mathon, R. B. Muniz, and M. S. Phan, *ibid.* **67**, 493 (1991); P. Bruno and C. Chappert, *ibid.* **67**, 1602 (1991); R. Coehoorn, Phys. Rev. B **44**, 9331 (1991).

[8] J. Ungaris, R. J. Celotta, and D. T. Pierce, Phys. Rev. Lett. **67**, 140 (1991).

[9] S. T. Purcell, W. Folkerts, M. T. Johnson, N. W. E. McGee, K. Jager, J. ann de Stegge, W. B. Zeper, W. Hoving, and P. Grünberg, Phys. Rev. Lett. **67**, 903 (1991).

[10] S. T. Purcell, M. T. Johnson, N. W. E. McGee, R. Coehoorn, and W. Hoving, "Two-Monolayer Oscillations in the Antiferromagnetic Exchange Coupling through Mn in Fe/Mn/Fe Sandwich Structures" (unpublished).

[11] A. Fuss, S. Demokritov, P. Grünberg, and W. Zinn, "Short- and Long-Period Oscillations in the Exchange Coupling of Fe across Epitaxially Grown Al and Au Interlayers" (to be published).

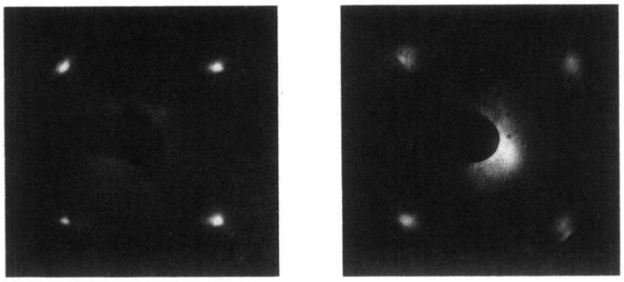
[12] Z. Q. Qiu, J. Pearson, and S. D. Bader, Phys. Rev. Lett. **67**, 1646 (1991).

[13] B. T. Jonker, J. Vac. Sci. Technol. A **8**, 3883 (1990).

[14] Z. Q. Qiu, J. Pearson, and S. D. Bader, "Additivity of the Magneto-opti Kerr Signal in Ultrathin Fe(110)/Ag(111) Superlattices" (to be published).

[15] J. Zak, E. R. Moog, C. Liu, and S. D. Bader, J. Magn. Magn. Mater. **89**, 107 (1990).

[16] P. M. Levy, J. L. Fry, and E. C. Ethridge, "Interlayer Magnetic Coupling in Transition-Metal Multilayered Structures" (unpublished).



(a) (b)

FIG. 1. The LEED patterns for (a) the Mo(100) surface, and (b) a Fe(14 ML)/Mo(n ML)/Fe(14 ML) sandwich.