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LETTER TO THE EDITOR

Oscillatory interlayer exchange coupling in magnetron sputtered NiFe/Mo multilayers, and comparison with Fe/Mo

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Abstract. NiFe (20 Å)/Mo multilayers were prepared by magnetron sputtering. We found typical oscillations of the interlayer coupling as a function of the Mo layer thickness with the same period as those in Fe/Mo multilayers (around 11 Å). However, the oscillation phases in NiFe/Mo and Fe/Mo are almost exactly opposite. The magnetoresistance measured at the first antiferromagnetic coupling state is very small.

Since antiferromagnetic (AFM) interlayer exchange coupling and giant magnetoresistance (GMR) were discovered in Fe/Cr sandwiches and superlattices [1, 2], much effort has been devoted to searching for various magnetic/non-magnetic metal multilayered systems that show AFM coupling and/or GMR. During the last few years, many multilayered systems have been found to belong to the same class of materials as Fe/Cr. Examples include Co/Cu [3], NiFe/Cu [4] and Fe/Mo [5, 6]. Generally, the nature of the interlayer exchange coupling in the magnetic multilayers is thought to arise from the spin polarization of the conduction electrons in the spacer layer adjacent to the magnetic layer. Various different approaches have been employed to model the interlayer exchange coupling, but they are often compared to the Ruderman–Kittle–Kasuya–Yosida (RKKY)-like behaviour with the result dependent upon the Fermi surface of the spacer materials. However, so far, rather extensive investigation of oscillatory interlayer exchange coupling has centred on its '*period*' and '*strength*', whereas little attention has been paid to the '*phase*'. In the early studies of the oscillatory interlayer exchange coupling, the phase shift had been observed in the Fe/Cu and the Co/Cu multilayers by Petroff *et al* [9]. In fact, different phases had already been seen by Parkin [10]. The phase shift of the interlayer exchange coupling in the ferromagnetic layer across the Cu(001) and Cu(110) has also been reported by Johnson *et al* [11]. In this paper, we report the interlayer exchange coupling in NiFe/Mo multilayers. The AFM interlayer exchange coupling and its oscillatory behaviour were observed. In comparison with Fe/Mo multilayers, we found that the oscillatory period is the same in NiFe/Mo and Fe/Mo, but the phases are almost exactly opposite. That is, we found the same period for multilayers with the same spacer material and different phases for multilayers with different ferromagnetic metals.

The [NiFe (20 Å)/Mo (t_{Mo})]$_{30}$ multilayers with variation of the Mo layer t_{Mo} were deposited on the water-cooled glass substrate by magnetron sputtering. The composition of

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the NiFe target was Ni 80% Fe 20%, and the material of the Mo target was 99.9% nominal purity. Samples were prepared in a working pressure of 0.5 Pa with the base pressure lower than 3×10^{-5} Pa. The deposition rates for NiFe and Mo were 2 and 1 Å s^{-1}, respectively. Without breaking the vacuum, a series of 16 samples with different multilayered structures was produced at a time, and the thickness of a single layer both for NiFe and for Mo was adjusted by controlling the deposition time via computerized control of the shutter and rotation of the substrate platform. The periodicity of the chemical composition modulation and the crystallographic structure of multilayers were confirmed by x-ray diffraction (XRD). The magnetic hysteresis loops of samples were measured at room temperature by using a vibrating sample magnetometer (VSM) and SQUID with the field in the film plane. The magnetoresistance was measured at room temperature by standard four-probe techniques with the magnetic field in the film plane but perpendicular to the sample current.

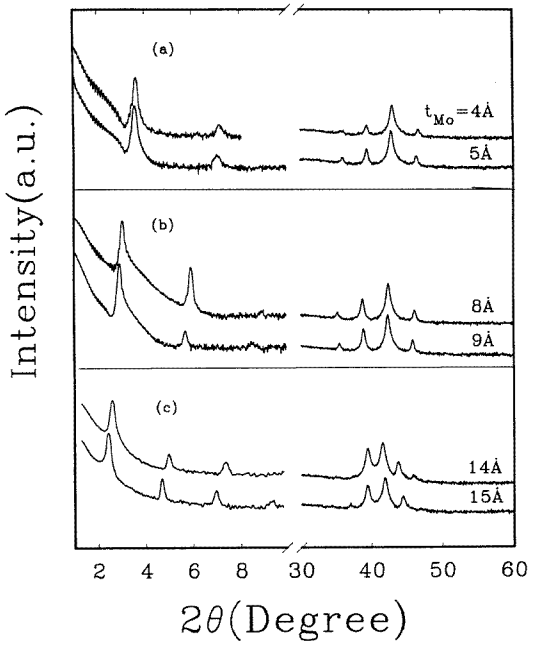

Figure 1. X-ray diffraction results for NiFe/Mo multilayers for a fixed Fe layer thickness of 20 Å. The data are plotted on a logarithmic intensity scale and results for each sample are shifted vertically for clarity. Left-hand panels are low-angle data and right-hand panels are high-angle data, respectively.

Low-angle x-ray diffraction revealed that superlattice peaks appear in the reflectivity spectra for all multilayered samples, even for ones with a small thickness of the Mo layer, indicating a well-defined chemical composition modulation along the growth direction. From the position of the superlattice peaks, the modulation periods were calculated. They agreed well with the designed values, indicating that good control of the fabrication process had been attained. The corresponding high-angle x-ray diffraction spectra for those samples show that the structure of the NiFe is FCC (111) texture, whereas that of the Mo layer is BCC (110) texture. Figure 1 shows examples of the low-angle and high-angle x-ray spectra of NiFe/Mo multilayers. The left-hand panels are low-angle diffraction data and the right-hand panels are high-angle x-ray diffraction data. The quality of the low-angle x-

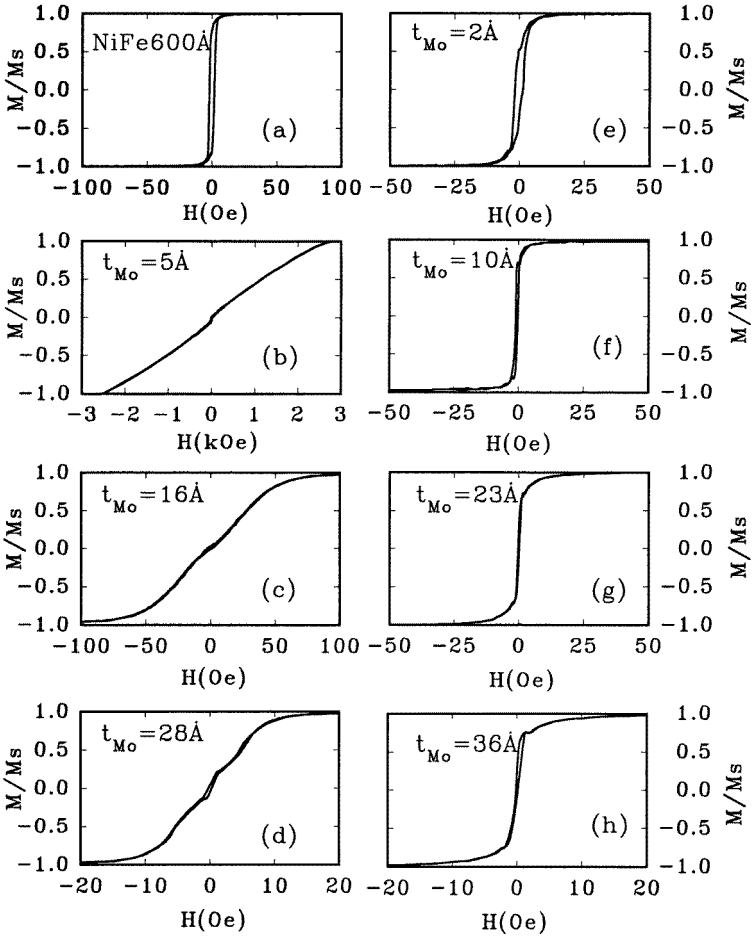

Figure 2. The room-temperature hysteresis loops are shown for eight representative samples: (a) is a hysteresis loop of a pure NiFe film of thickness 600 Å ($t_{Mo} = 0$ Å); (b), (c) and (d) are for films that are antiferromagnetically coupled with Mo layer thicknesses $t_{Mo} = 5$, 16 and 28 Å, respectively; (e), (f), (g) and (h) are for films that are ferromagnetically coupled with Mo layer thicknesses $t_{Mo} = 2$, 10, 23 and 36 Å, respectively.

ray diffraction spectra indicates that the films are well layered, similar to Fe/Mo multilayers [5]. The corresponding high-angle x-ray diffraction spectra, as expected, yield satellite peaks around primary Bragg peaks, indicting a high degree of order. Our structure refinement of the NiFe/Mo multilayers from x-ray diffraction data will be presented in more detail in a further publication.

Figure 2 shows the representative magnetization behaviours of NiFe/Mo multilayers at room temperature. It is seen in figure 2(a) that the hysteresis loop of a pure NiFe film of thickness 600 Å, which exhibits a larger remanence M_r and very small saturation field H_s, is fully ferromagnetic. Similarly, the hysteresis loops in figures 2(e), (f), (g) and (h) for $t_{Mo} = 2$, 10, 23 and 36 Å have larger remanence M_r and small H_s, also indicating that neighbouring magnetic layers of those samples are ferromagnetically coupled. However, loops (b), (c) and (d) with $t_{Mo} = 5$, 16 and 28 Å are very different from loops (a), (e), (f),

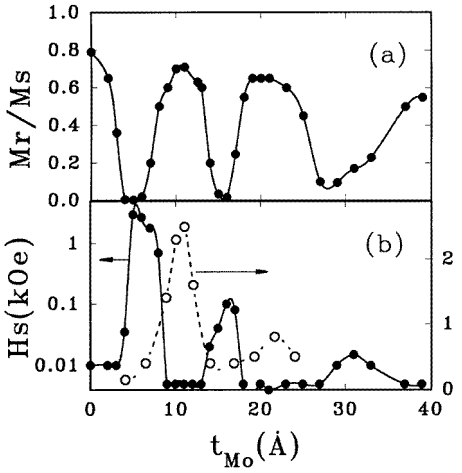

Figure 3. The variation of (a) M_r/M_s, and (b) H_s as a function of the Mo layer thickness for NiFe/Mo multilayers. The variation in H_s as a function of the Mo layer thickness for Fe/Mo multilayers is also shown in (b) (open symbols). The full and broken lines are guides for the eye. Note the different vertical scales for NiFe/Mo and Fe/Mo.

(g) and (h). The remanence M_r of the loops is nearly reduced to zero and the saturation field H_s of the loops is large. Notice the different scale on the horizontal ordinates. Clearly, the magnetization behaviour of those samples is typical of AFM exchange coupling. We estimated the AFM exchange coupling strength of NiFe/Mo multilayers in a way similar to that in [12], calculating the exchange coupling strength $J = M_s\ t_{NiFe}\ H_s/4$ to yield $J = 0.020$ erg cm^{-2} at $t_{Mo} = 5$ Å, which is much smaller than those of Fe/Cr [12] and Fe/Mo [6].

Figure 3 shows the variation of (a) M_r/M_s, and (b) H_s as a function of the thickness of the Mo layer. M_r/M_s of the pure NiFe single film of thickness 600 Å is 0.8. In the NiFe/Mo multilayer, as the thickness of the Mo layer increases, the M_r/M_s ratio decreases to its first minimum value (0.006) at $t_{Mo} = 5$ Å. This value near to zero means that the AFM exchange coupling at this thickness is almost perfect. As the Mo layer thickness increases further, the M_r/M_s value increases, reaching its maximum value at $t_{Mo} = 10$ Å. A further oscillation in the M_r/M_s ratio is clearly observed for even larger values of t_{Mo} with its minimum values at $t_{Mo} = 16$ and 28 Å. Those results show that the NiFe/Mo system exhibits the oscillatory behaviour typical of interlayer exchange coupling. The period of oscillation is about 11 Å, which is equal to that of Fe/Mo multilayers [5, 6]. The saturation field H_s in figure 3(b) is also seen to be oscillatory, its maximum values appearing at $t_{Mo} = 5$, 16 and 28 Å, corresponding to the AFM state. Note that the envelope of the maxima of the H_s value reduced rapidly in successive AFM states. This result suggests that the AFM coupling strength falls off rapidly as the thickness of the Mo layer increases. However, the magnetoresistance of the sample at the AFM peak is extremely small. For the sample at the first AFM exchange coupling state with a 5 Å Mo layer, the magnetoresistance ratio $\Delta R/R$ is only about 0.3% at room temperature. The reason for such a small magnetoresistance may be related to the high resistivity of the NiFe/Mo multilayers, namely the reduction in the mean free path of conducting electrons.

The most striking result is the larger phase shift of the oscillatory interlayer exchange coupling between NiFe/Mo and Fe/Mo multilayers. Figure 3(b) shows H_s as a function

of the Mo layer thickness both for the NiFe/Mo and for the Fe/Mo system. It can be seen in figure 3(b) that the first AFM state in the NiFe/Mo system appears for 5 Å Mo layer thickness; the second and third AFM states appear at 16 and 28 Å, respectively. For the Fe/Mo system, however, the first AFM state appears at 10 Å Mo layer thickness and the second AFM state appears at 22 Å. It is evident that the interlayer exchange coupling between NiFe layers is AFM in the thickness ranges of the Mo layer within which the exchange coupling between Fe layers is ferromagnetic (FM) and vice versa. A similar observation has been reported for Fe/Cu and Co/Cu systems by Petroff *et al* [9], but no further analysis was given. The dependence of the phase of the oscillatory exchange coupling between ferromagnetic layerd across FCC Cu on the composition of the ferromagnetic layer was reported recently by Johnson *et al* [11]. They observed the variation of the phase shift in the oscillatory interlayer exchange coupling across Cu(001) and Cu(110). In the case of coupling across Cu(001), the phase of the short-period oscillation varies monotonically with the number of d electrons in the FM layer, whereas the phase of long-period oscillation remains constant. In contrast, in the case of (110) oriented systems the phase varies conspicuously non-monotonically with the d band occupation of the magnetic layers. The band gaps at crucial points of the ferromagnets' spin-dependent Fermi surface, within Bruno's 'electron optics' model for interlayer exchange coupling, are thought to be responsible for the shift in the phase. The numerical studies by Mathon [13] showed that the oscillation phase depends strongly on the matching between the band in the spacer and magnetic elements, which is similar to Bruno's model. However, the phase shifts obtained for the different theoretical models are different, which indicates that the phase is a model-dependent quantity. Obviously, more studies are needed in order to understand the mechanisms for the interlayer exchange coupling and the relationship of the phase shift to it better. We hope that our experimental results could give the theorists additional assistance in their analysis.

In summary, we have prepared NiFe/Mo multilayers by magnetron sputtering. The multilayers exhibit a typical interlayer exchange coupling oscillation with a period of 11 Å. In comparison with Fe/Mo, the oscillation phase is almost exactly the opposite. Clearly, the phase's dependence on different magnetic elements with the same non-magnetic spacer needs to be investigated further. In addition, only a small negative magnetoresistive anomaly was observed in the AFM exchange coupling state. The small MR value may be governed by the high resistivity that reduces greatly the mean free path of conduction electrons.

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