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*Work supported by the U.S. Department of Energy, Basic Energy Sciences-Materials Sciences, under contract #W-31-109-ENG-38.

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Structural and magnetic properties of sputtered Ni/Mo superlattices were studied for Ni thicknesses of 14 and 30 Å and Mo thicknesses from 3 to 25 Å. The sign of the interlayer magnetic coupling oscillates between ferromagnetic and antiferromagnetic (AF) with a period of ≈ 12 Å of Mo thickness. The oscillation is similar in phase to that for Co/Mo superlattices, but is shifted out of phase from that published for Fe/Mo superlattices, in accord with theoretical expectation. The temperature dependence of the saturation magnetization below ~ 150 K follows a $T^{3/2}$ -behavior for the 30-Å Ni films and is quasi-linear, $T \ln T$, for the 14-Å films, which indicates a dimensionality crossover from 3D to 2D, respectively.

PACS# 75.70.-i, 75.70.Fr, 68.65.+g

* Work supported by the U.S. Department of Energy, BES-Materials Sciences, under contract W-31-109-ENG-38.

Ni/Mo was among the first examples of metallic heterostructures that were found to grow as high-quality superlattices.¹ As a result, there is a significant body of work in the literature that examines its structural,¹⁻³ elastic,^{1,4} transport^{1,5} and magnetic properties.^{6,7} Most recently considerable interest has focused on the indirect exchange coupling of magnetic layers interleaved with nonmagnetic metals in superlattice structures.⁸⁻¹¹ Oscillations in the sign of the interlayer coupling have been observed for a wide variety of nonmagnetic spacer materials.⁸ However, no prior study, to our knowledge, has been devoted to the interlayer magnetic coupling of the Ni/Mo system. In this Letter, we report structural and magnetic properties of sputtered Ni/Mo superlattices and compare the results to similar studies of Fe/Mo¹⁰ and Co/Mo^{8,12} superlattices. We find that Ni/Mo superlattices possess an interlayer coupling which oscillates with a phase and period close to that found for Co/Mo, but out of phase with that reported for Fe/Mo superlattices, in agreement with recent predictions^{13,14} We also find that at low temperature the magnetization (and the interfacial coupling energy) follow a two-dimensional $T \ln T$ law for superlattices with thin Ni layers, as recently predicted.¹⁵ Our work is based on series of Ni/Mo superlattices grown by d.c. magnetron sputtering, as described in Ref. 10. The Mo-layer thicknesses range from 3-25Å and the Ni-layer thicknesses are 14 and 30Å. We characterized the structure with x-ray diffraction (XRD) and the magnetic properties by SQUID magnetometry.

Both low-angle and crystalline high-angle XRD superlattice peaks were observed for all Mo thicknesses, indicative of good chemical layering and structural coherence. XRD spectra for a $[\text{Ni}(14\text{\AA})/\text{Mo}(19\text{\AA})]_{20}$ superlattice are shown in Fig. 1. To determine the out-of-plane crystal structure, the high-angle spectrum was refined to a general structural model.^{3,16,17} The result is shown by the solid line in Fig. 1a. The spectrum could not successfully be refined by assuming that the strain profile of the Mo-on-Ni interface is equivalent to that of the Ni-on-Mo interface. This is consistent with the results of Ref. 3, where asymmetric strains in the Ni layers were found. Shown in the inset of Fig. 1a is the average Mo (squares) and Ni (circles) lattice spacings in the growth direction for the $[\text{Ni}(14\text{\AA})/\text{Mo}]_{20}$ series. The average Ni lattice spacing is expanded over the bulk value by $\approx 3\%$ for all Mo thicknesses. The Mo layer is contracted relative to its bulk value,

with the magnitude depending strongly on Mo thickness. Figure 1b shows four low-angle Bragg reflections, and higher frequency finite-size oscillations indicative of smooth layers with limited cumulative roughness.¹⁸ The low-angle spectrum was fitted to a general optical formalism¹⁹ (solid line in Fig. 1b) in which the interfacial profiles were assumed to be of Gaussian width σ . The fitted value for σ of 2.5Å characterizes the interfacial roughness and/or chemical interdiffusion. The layer-thickness values are consistent with those obtained from the high-angle data to better than 1Å.

Selected samples were removed from their substrates and transmission XRD was performed to determine the in-plane structure. The samples are polycrystalline in-plane. The transmission results for a Ni(11Å)/Mo(11Å) superlattice show that the Mo layers asymmetrically strain in-plane by -2.6% along the [002] and -0.6% along the [1 $\bar{1}$ 0], and that the Ni layer expands $\approx$ 1.6% along the [2 $\bar{2}$ 0] direction. The strain anisotropy is due to the large difference in lattice-mismatch along orthogonal directions, and is consistent with studies of a number of other fcc/bcc superlattices.^{2,20,21} For thicker layers, the lattice strains relax and bulk spacings are approached.

The results for the thickness dependence of the saturation magnetization (M_S) and the Curie temperature (T_C) are in quantitative agreement with previous studies.⁶ The low-temperature ($T=5K$) moment of the $t_{Ni}=14Å$ and $30Å$ films are 125 ± 20 and 310 ± 20 emu/cc, respectively. These values are significantly reduced from the bulk value of 522 emu/cc. SQUID measurements give an average magnetization; without a measurement of the magnetization profile, we cannot differentiate between changes in the average moments vs. interfacial moments.

$M_S(T)$ for [Ni(30Å)/Mo(12Å)]₂₀ and [Ni(14Å)/Mo(12Å)]₂₀ superlattices are shown in Fig. 2. The $t_{Ni}=14Å$ films have low- T_C values (near 200K) and the $t_{Ni}=30Å$ films order well above room temperature. We fitted the $t_{Ni}=30Å$ M_S -vs- T data to a standard power-law expression:

$$M_S(T) = M_S(0) [1 - a T^\alpha], \quad (1)$$

as shown by the solid line in Fig. 2a. The 30Å Ni films follow the expected 3D $T^{3/2}$ behavior. The exponent is insensitive to the T range selected for the fit, and varies by only 0.05 when the upper T -range of the fit is increased from 75 to 225K. The temperature dependence of M_S for the

$t_{\text{Ni}}=14\text{\AA}$ sample is qualitatively different, as seen in Fig. 2b. In this case $M_S(T)$ is best fitted by the quasi-linear relation:

$$M_S(T) = M_S(0) [1 - b T \ln T], \quad (2)$$

which is the expected spin-wave behavior for a 2D magnetic layer.^{15,22}

The Ni interlayer coupling was determined from the saturation field (H_S) of the magnetic hysteresis loops. Shown in Fig. 3 are the low field regions measured at $T=50\text{K}$ for the $[\text{Ni}(14\text{\AA})/\text{Mo}]_{20}$ superlattices with $t_{\text{Mo}}=7, 12$ and $19\text{\AA}$. The $t_{\text{Mo}}=7$ - and $19\text{\AA}$ samples show the characteristic features of AF coupling, a large H_S value and a magnetization plateau at low fields. The intermediate Mo thickness ($t_{\text{Mo}}=12\text{\AA}$) sample has a square hysteresis loop indicative of either uncoupled or ferromagnetically coupled Ni layers. H_S vs Mo thickness is plotted in Fig. 3b. H_S is peaked at $t_{\text{Mo}}=6$ and $18\text{\AA}$ which yields a periodicity of $\approx 12\text{\AA}$ in agreement with the $11\text{\AA}$ period found for both Fe/Mo ¹⁰ and Co/Mo ^{8,12} superlattices. The phase of the oscillation agrees with that for Co/Mo , but is out of phase with respect to Fe/Mo where the first AF peak appears at $t_{\text{Mo}}=12\text{\AA}$. This result also agrees with the recent theoretical work,^{13,14} which predicts that the AF coupling in Co- and Ni-based superlattices should oscillate with similar phase but different from that in Fe-based superlattices.

The temperature dependence of the AF-coupling energetics was studied for the $14\text{\AA}$ Ni superlattice. Figure 4a contains hysteresis loops for $[\text{Ni}(14\text{\AA})/\text{Mo}(7\text{\AA})]_{20}$ that show that the M_S values change by a factor of four between 25 and 150K, due to the low T_C value. Figure 4b shows the same data normalized to M_S . *The shape of the hysteresis loops are nearly identical with no measurable change in H_S value.* Since the interfacial coupling energy is approximately equal to $H_S M_S t_{\text{Ni}}/4$, the constancy of H_S implies that the coupling energy possesses the same $T \ln T$ dependence as M_S over this temperature range, in agreement with recent theoretical analysis by Cullen and Hathaway.¹⁵ In comparing the coupling energies of the $t_{\text{Ni}}=14$ and $30\text{\AA}$ samples, we find that the product $H_S t_{\text{Ni}}$ remains constant. This confirms that the coupling energy scales with M_S even though M_S has increased by $\sim 250\%$ for the thicker Ni layers. (Similar measurements of the dependence of the coupling energy and M_S on T for the second AF-coupling peak were

hampered by the strong temperature dependence of the coercive field H_C . For low temperatures, the value of H_C becomes comparable to that of H_S and small changes in H_S are difficult to resolve. Nonetheless, the changes in H_S are significantly smaller than those in M_S .)

In summary, we presented structural and magnetic properties of sputtered Ni/Mo superlattices. We find the AF coupling oscillates with a period of $\approx 12\text{\AA}$. The phase of the oscillation is similar to that for Co/Mo superlattices, but is out of phase with that observed in Fe/Mo superlattices. The temperature dependence of M_S exhibits a crossover from 2D-to-3D behavior as a function of increasing Ni thickness. Both M_S and the AF coupling energy for the 14- $\text{\AA}$ Ni possess the predicted $T \ln T$ behavior at low temperature, while thicker Ni films follow the $T^{3/2}$ Bloch law.

Acknowledgment

Work supported by the U.S. Department of Energy, BES-Materials Sciences, under contract W-31-109-ENG-38.

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Figure 1: (a) High-angle and (b) low-angle x-ray diffraction result for a $[\text{Ni}(14\text{\AA})/\text{Mo}(19\text{\AA})]_{20}$ superlattice. Open circles are measured intensities and the solid line represents the best fit. The inset shows average Mo (squares) and Ni (circles) lattice spacing in the growth direction determined by fitting the high-angle spectra of $[\text{Ni}(14\text{\AA})/\text{Mo}(t_{\text{Mo}})]_{20}$ superlattices.

Figure 2: Open squares are the measured $M_S(T)$ values for (a) $[\text{Ni}(30\text{\AA})/\text{Mo}(12\text{\AA})]_{20}$ and (b) $[\text{Ni}(14\text{\AA})/\text{Mo}(12\text{\AA})]_{20}$ superlattices. Solid lines are least-squares fits to (a) power law and (b) quasi-linear $T \ln T$ behaviors.

Figure 3: (a) Magnetic hysteresis loops for $[\text{Ni}(14\text{\AA})/\text{Mo}(t_{\text{Mo}})]_{20}$ superlattices measured at 50K. (b) H_s vs. t_{Mo} for $[\text{Ni}(14\text{\AA})/\text{Mo}(t_{\text{Mo}})]_{20}$ superlattices.

Figure 4: Magnetic hysteresis loops for a $[\text{Ni}(14\text{\AA})/\text{Mo}(7\text{\AA})]_{20}$ superlattice measured at 25, 50, 100 and 150K, where (a) shows the absolute magnetization; and (b) shows the normalized magnetization of only half the measured loops for clarity.

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Figure 1