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# Oscillations in Exchange Coupling and Magnetoresistance in Metallic Superlattice Structures: Co/Ru, Co/Cr, and Fe/Cr

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We report the discovery of antiferromagnetic interlayer exchange coupling and enhanced saturation magnetoresistance in two new metallic superlattice systems, Co/Cr and Co/Ru. In these systems and in Fe/Cr superlattices both the magnitude of the interlayer magnetic exchange coupling and the saturation magnetoresistance are found to oscillate with the Cr or Ru spacer layer thickness with a period ranging from 12 Å in Co/Ru to ≃18–21 Å in the Fe/Cr and Co/Cr systems.

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Recently, it was reported that single-crystal multilayered structures containing Fe layers coupled through thin (≃5–20 A) Cr layers display unexpectedly high values of saturation magnetoresistance.[1,2] The change in resistance is closely related to changes in the magnetic structure of the superlattice with magnetic field. In zero field, the magnetic moments of Fe layers on either side of the Cr spacer layer are ordered antiparallel to one another.[3,4] On application of a sufficiently large magnetic field the moments become aligned in the same direction and the magnetoresistance saturates. In this Letter, we report that the saturation magnetoresistance in Fe/Cr superlattice structures does not decrease monotonically with increasing Cr layer thickness as was previously reported,[1] but oscillates in magnitude as a function of the Cr layer thickness. In addition, we report similar behavior in two other systems, Co/Cr and Co/Ru. Furthermore, we show that in all three systems the magnitude of the saturation field oscillates with the same period as that of the saturation magnetoresistance. To our knowledge these are the first reported observations of such effects in transition metals other than the heavy rare-earth metals.

The superlattice structures for these studies were deposited on chemically etched Si(111) wafers in a high-vacuum dc magnetron sputtering system containing four magnetron sources. The base pressure of the vacuum system prior to deposition was better than $\simeq 2\times 10^{-9}$ Torr. The structures were prepared in 3.2 mTorr of argon at a deposition rate of 2 Å/sec with substrate temperatures varying from 40 to 450 °C. A series of up to nineteen samples of arbitrarily complex structures was prepared without breaking vacuum *via* computerized control of the substrate platform and shutters located between each magnetron source and the platform.

Auger sputter depth profiling, using Zalar rotation, of several samples showed no significant amounts (< 1%) of oxygen or carbon in the metal layers, and confirmed that the sequence of layers and their relative thicknesses were as programmed. The structures of numerous samples were examined in more detail using a standard $\theta/2\theta$ x-ray powder diffractometer. Both low- and high-angle superlattice diffraction peaks were observed. The superlattice wavelength determined from the position of these peaks was typically within 10% of that inferred from the thickness of calibration films, measured using a surface profilometer, deposited with the superlattice structures. In addition, the x-ray data established that the samples were polycrystalline with weak texturing, and that the Fe and Cr layers in Fe/Cr were bcc and the Co and Ru layers in Co/Ru hcp. The line widths of the diffraction peaks indicated structural coherence lengths of ≃150 Å. Through-foil transmission-electron-microscopy studies on samples specially deposited on single-crystal NaCl and prethinned SiN wafers showed grain sizes of approximately the same size 100–200 Å.

A number of samples were examined by cross-section transmission electron microscopy (XTEM). Figure 1 shows a high-resolution micrograph taken along the Si $[11\bar{2}]$ direction of the structure Si(111)/(100 Å) Ru/

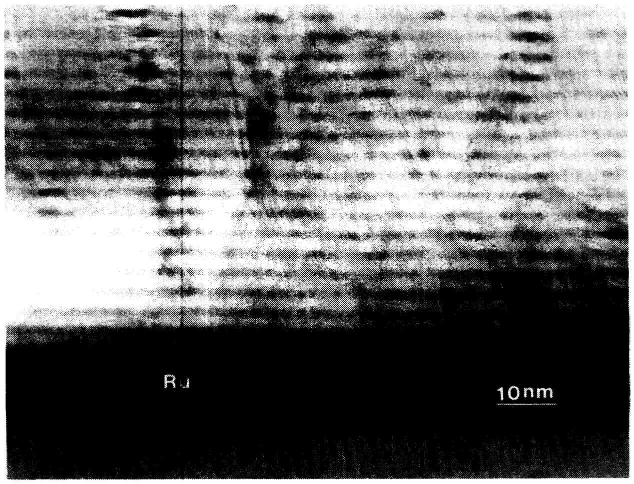

FIG. 1. High-resolution micrograph taken along the Si $[11\bar{2}]$ direction from the structure, Si(111)/(100 Å) Ru/[(18 Å) Co/(8 Å) Ru$]_{20}$/(50 Å) Ru. The sample was deposited at 40 °C.

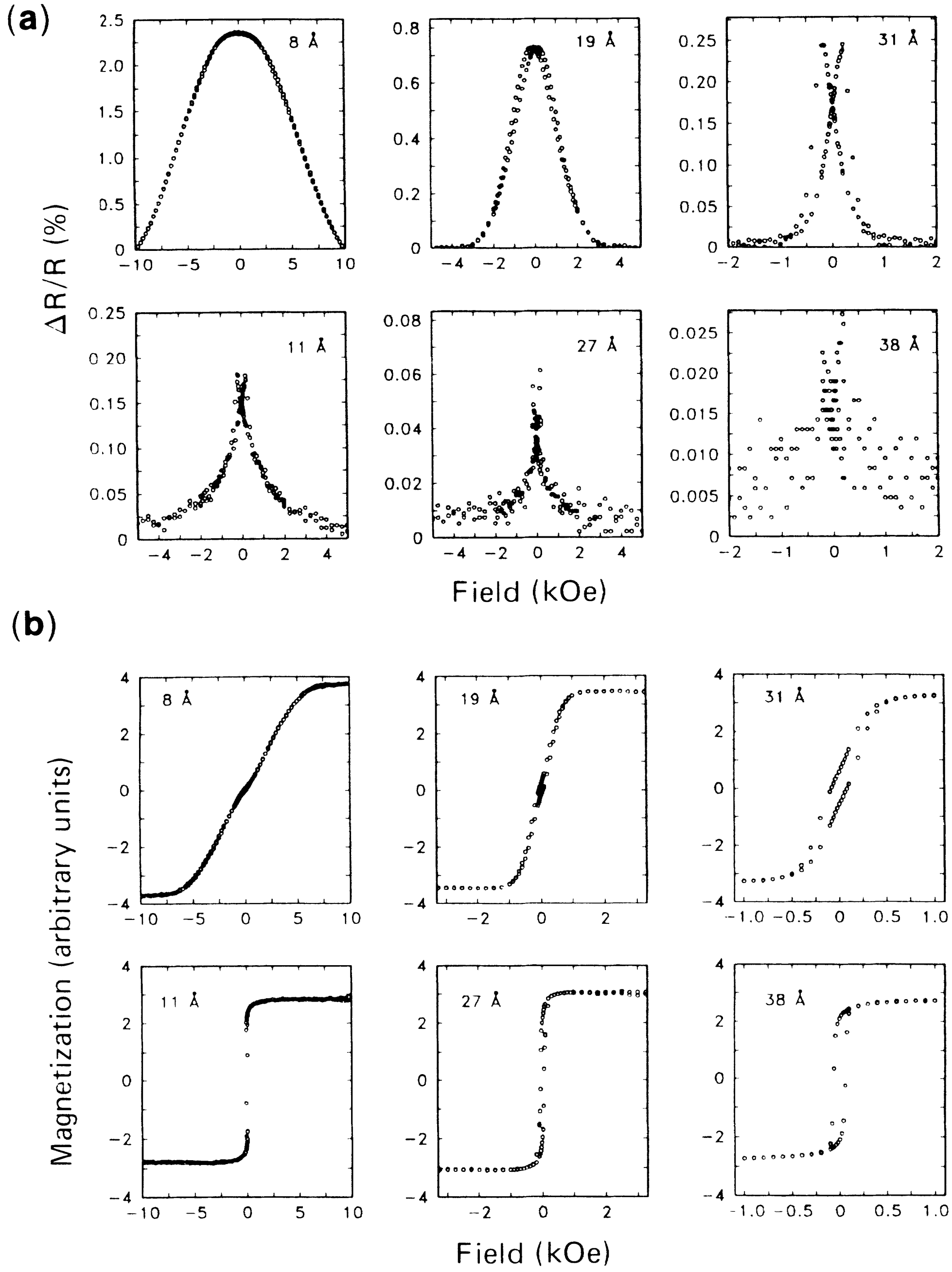

FIG. 2. Typical (a) transverse magnetoresistance (4.5 K) and (b) magnetization (300 K) vs in-plane field curves for six Co/Ru superlattice structures deposited at 125 °C. The structures are of the form Si(111)/(100 Å) Ru/[(18 Å) Co/$t_{\mathrm{Ru}}$ Ru]$_{20}$/(50 Å) Ru with Ru layer thicknesses $t_{\mathrm{Ru}}$ of 8, 11, 19, 27, 31, and 38 Å.

[(18 Å) Co/(8 Å) Ru]$_{20}$/(50 Å) Ru. The micrograph shows that the superlattice has high structural perfection with surprisingly flat interfaces. The layers are contiguous over lengths greater than 0.25 μm with no evidence for disruption of these layers. In addition, the micrograph reveals that the structure is comprised of grains of the order of 150 Å in size, i.e., several bilayers, within which lattice fringes are observed which are coherent across the Co/Ru interfaces. Similar micrographs on Fe/Cr superlattice structures show much less perfect structures with rumpled disconnected layers.[5]

Resistance and magnetization[6] versus in-plane magnetic-field curves are shown in Fig. 2 for six Co/Ru superlattice structures containing 18-Å-thick Co layers and Ru layer thicknesses ranging from 8 to 38 Å. Figure 3 shows the detailed dependence of the saturation mag-

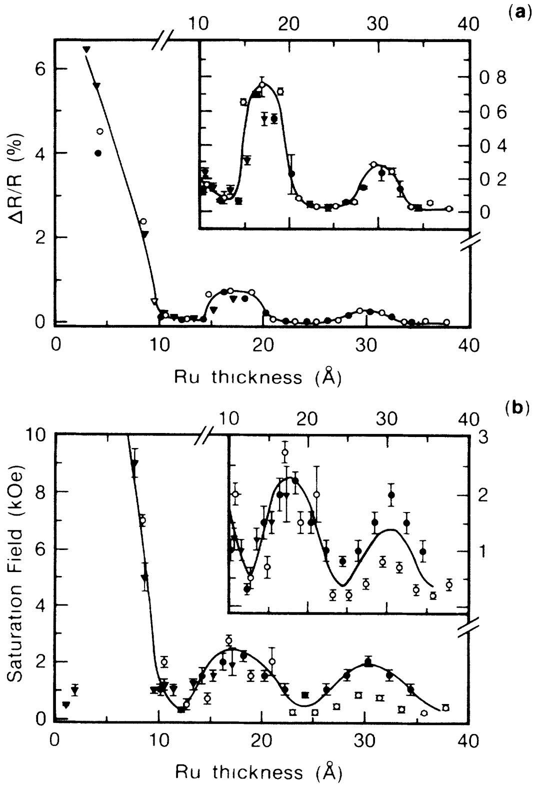

FIG. 3. (a) Transverse saturation magnetoresistance (4.5 K) and (b) saturation field (300 K) vs Ru layer thickness for structures of the form Si(111)/(100 Å) Ru/[20 Å) Co/$t_{\mathrm{Ru}}$ Ru]$_{20}$/(50 Å)Ru deposited at temperatures of ●, 40 °C; ○, 125 °C; ×, 200 °C.

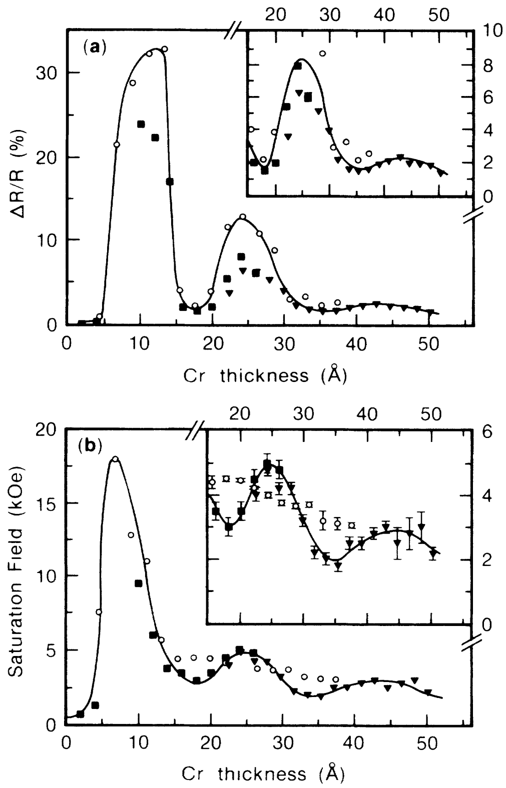

FIG. 4. (a) Transverse saturation magnetoresistance (4.5 K) and (b) saturation field (4.5 K) vs Cr layer thickness for three series of structures of the form Si(111)/(100 Å) Cr/[(20 Å) Fe/$t_{\mathrm{Cr}}$ Cr]$_N$/(50 Å) Cr, deposited at temperatures of △,■, 40 °C ($N=30$); ○,125 °C ($N=20$).

netoresistance and the saturation field on the thickness of the Ru layer. For $t_{\mathrm{Ru}}=8$ Å the field dependence of resistance and magnetization are similar to that for comparable Fe/Cr structures, but the magnitude of $\Delta R/R$ (Ref. 6) is much smaller. $\Delta R/R$ and the saturation field[6] $H_s$ increase as $t_{\mathrm{Ru}}$ is decreased below $\simeq 8$ Å, attaining values of 6.5% and $>70$ kOe, respectively, for $t_{\mathrm{Ru}} \simeq 3$ Å or approximately 2 monolayers. The saturation field reflects the strength of the antiferromagnetic interlayer exchange coupling $J_i$ as $-4J_i=H_s M t_F$,[7] where $M$ and $t_F$ are the magnetization and the thickness of the ferromagnetic layer, respectively. For this film, $J_i \simeq -5$ ergs/cm$^2$, a very large value which is almost 2-3 times larger than the largest antiferromagnetic exchange-coupling values found in Fe/Cr structures. These values are so large that they clearly cannot be accounted for by magnetostatic coupling of the ferromagnetic layers.

As $t_{\mathrm{Ru}}$ is increased beyond 8 Å both $\Delta R/R$ and $H_s$ decrease to very small values for $t_{\mathrm{Ru}}$ in the range 10-14 Å. As $t_{\mathrm{Ru}}$ is increased further both $\Delta R/R$ and $H_s$ become larger again reaching maximum values for $t_{\mathrm{Ru}}$ close to 18 Å. These values are much smaller than those found for very thin Ru layers. A further oscillation in these quantities is observed for even larger values of $t_{\mathrm{Ru}}$ with peak values for $t_{\mathrm{Ru}} \simeq 31$ Å. These data unambiguously demonstrate that the saturation field and saturation magnetoresistance are closely related and that their magnitude oscillates as a function of $t_{\mathrm{Ru}}$ with a period of approximately 12-14 Å.

Data for Fe/Cr superlattices for Cr thicknesses in the range 10-20 Å are reported elsewhere.[8] Both the transport and magnetic properties of these sputtered polycrystalline Fe/Cr superlattice structures are similar to those reported for molecular-beam epitaxy grown single-crystal structures[1] with comparably large maximum values of $\Delta R/R$ and $J_i$ of $\simeq 50\%$ and 1.0 erg/cm$^2$, respectively. However, in contrast to the Co/Ru system, values of $\Delta R/R$ depend sensitively on deposition conditions and can easily vary by a factor of 2 for a given structure. This makes the determination of the thickness dependence of $\Delta R/R$ difficult unless large numbers of samples can be prepared simultaneously under similar deposition conditions. Figure 4 shows the dependence of $\Delta R/R$ and $H_s$ on the Cr layer thickness $t_{\mathrm{Cr}}$ for several series of structures of the form Si(111)/(100 Å) Cr/[(20 Å) Fe/$t_{\mathrm{Cr}}$ Cr]$_N$/(50 Å) Cr ($N=20$ or 30). Although the magnitude of $\Delta R/R$ is much larger for films deposited at

125 °C than for films deposited at 40 °C, at both temperatures well-defined oscillations are found in both the saturation magnetoresistance and the saturation field —determined from the resistance curves—as a function of Cr layer thickness, as shown in the figure. Peaks in $\Delta R/R$ are found at values of $t_{\mathrm{Cr}}$ of approximately 9, 25, and 43 Å with peaks in $H_s$ at $t_{\mathrm{Cr}}$ of 7, 24, and 43 Å giving an oscillation period of $\simeq 18$ Å. The oscillation period is significantly larger than that for Co/Ru, and, moreover, the peaks and valleys are systematically shifted by approximately half an oscillation period compared to the Co/Ru system. This means that whereas for the Fe/Cr system $H_s$ and $\Delta R/R$ decrease as $t_{\mathrm{Cr}}$ is decreased below $\simeq 8$ Å, for the Co/Ru structures these quantities are becoming progressively larger as the Ru layer thickness is decreased down to $\simeq 3$ Å.

Similar studies were made on a large number of other superlattice systems: In particular, it was found that Co/Cr superlattice structures exhibit behavior similar to Co/Ru with a larger oscillation period of about 21 Å but with still smaller values of $\Delta R/R$ and $H_s$. The largest values of $\Delta R/R$ and $H_s$ found were 2.5% and 6 kOe, respectively, for a $[(15\ \text{Å})\ \mathrm{Co}/(4\ \text{Å})\ \mathrm{Cr}]_{30}$ structure.

The oscillatory dependence of magnetoresistance and exchange coupling with spacer-layer thickness in three different metallic superlattice structures suggests that this is a common feature of such systems. One possible origin of the exchange coupling suggested by this work is some sort of Ruderman-Kittel-Kasuya-Yosida[9] (RKKY) coupling mediated by spin polarization of the Cr or Ru layers. However, such a mechanism gives rise to ferromagnetic coupling as the distance between the magnetic moments becomes very small which appears to be in contradiction with the results for the Co/Ru structures.[10] In addition, since the period of the RKKY oscillation is tied to the Fermi wave vector, a much shorter oscillation period than that found of just 2–3 lattice spacings would be expected, such as that postulated, for example, in certain spin-glass systems.[11] Since Ru is nonmagnetic a mechanism based on a long-range-ordered magnetic state of the spacer layer seems to be ruled out. Indeed, for the Fe/Cr system, enhanced $\Delta R/R$ and large $H_s$ are observed well above the bulk Néel temperature of Cr (312 K). One might speculate that the ordering temperature of thin Cr layers is substantially different from that of the bulk material, but since we find that the antiferromagnetic exchange coupling persists up to temperatures of $\simeq 625$ K, diminished in magnitude by only $\simeq 30\%$ compared to 4.2 K, this seems unlikely. For the Co/Ru system a larger temperature dependence of $J_i$ is found, but again large values of $J_i$ are found for temperatures up to 625 K.

In conclusion, we have observed oscillations, as a function of spacer-layer thickness, in the magnitude of saturation magnetoresistance and the magnetic exchange coupling in three different metallic superlattice systems, Co/Ru, Fe/Cr, and Co/Cr. The period of the oscillations ranges from $\simeq 12$ to 21 Å. The newly discovered Co/Ru system is particularly interesting in that the spacer layer is nonmagnetic. Moreover, the interlayer exchange-coupling energy is much greater than that found in the Fe/Cr system. For both Co/Cr and Co/Ru, in distinct contrast to Fe/Cr, the exchange coupling is *antiferromagnetic* in the limit of ultrathin continuous spacer layers. It seems to be difficult to reconcile this result with a RKKY coupling mechanism suggested by the oscillatory dependence of exchange coupling on the magnetic layer separation.

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[6] The resistance of the samples was measured using a four-probe low-frequency ac lock-in technique with spring-loaded pressure contacts. All the magnetoresistance data presented in this paper were measured at 4.5 K. The saturation magnetoresistance $\Delta R/R$ is defined as the ratio of the difference between the peak in resistance of the sample at low field and the resistance at high field divided by the value at high field. For samples with thick spacer layers of Cr or Ru the peak of the resistance curve at low fields was shifted from zero field and displayed hysteresis. Magnetization data were measured using vibration sample and SQUID magnetometry. The saturation field $H_s$ was defined as the field at which the resistance or magnetization curve first deviates from the high-field slope.

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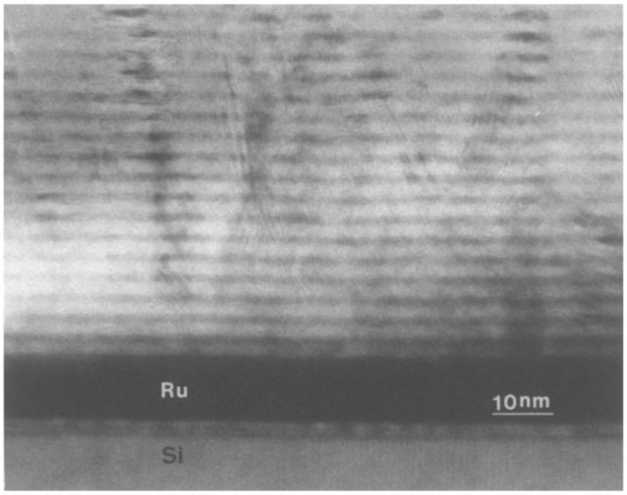
[10] Note that while data are included in Fig. 3 for a Ru layer thickness of 1 Å we assume that at this thickness the Ru layer is discontinuous so that the Co layers are directly exchange coupled.

[11] See, for example, K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, Amsterdam, 1984).



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