Systematic Variation of the Strength and Oscillation Period of Indirect Magnetic Exchange Coupling through the 3*d*, 4*d*, and 5*d* Transition Metals

S. S. P. Parkin

IBM Research Division, Almaden Research Center, 650 Harry Road, San Jose, California 95120-6099

(Received 26 April 1991)

We show that oscillatory indirect magnetic exchange coupling via transition metals sandwiched between ferromagnetic layers of Fe, Co, Ni, or Ni alloys is a general phenomenon. Surprisingly, the oscillation period is approximately the same, $\simeq 10$ Å, in all the transition metals in which we observe coupling with the single exception of Cr, for which it is much longer. Furthermore, the exchange-coupling strength is found to increase systematically from the 5*d* to 4*d* to 3*d* metals and *exponentially* with increasing number of *d* electrons along each period.

PACS numbers: 75.50.Rr, 72.15.Gd, 75.70.Cn

The discovery [1,2] of long-range oscillatory indirect magnetic exchange coupling between two ferromagnetic layers separated by thin layers of the nonmagnetic transition metals, Cr, Ru, and Cu, has stimulated considerable interest [3–11]. Prior to this work, oscillatory indirect exchange coupling via transition metals (TM) had been inferred only from studies on dilute magnetic alloys (see, for example, Ref. [12]). In RKKY [13,14] models used to account for the properties of such alloys, the conduction electrons become spin polarized in the immediate neighborhood of a magnetic impurity. The spin polarization, and consequently the coupling to nearby magnetic impurities, decays with increasing distance from the magnetic impurity in an oscillatory manner. The oscillation period is given for a free-electron gas by half the Fermi wavelength, $\lambda_F/2$, of the host nonmagnetic metal, which is quite short at just $\simeq 1$–2 atomic spacings. In contrast the magnetic coupling through thin layers of Cr, Ru, and Cu oscillates with much longer periods, P_{TM}, ranging from $P_{Cu} \simeq 10$ Å [2,6,7] and $P_{Ru} \simeq 11$ Å [1] to $P_{Cr} \simeq 18$–20 Å [1].

The transition metals clearly do not have simple free-electron-like Fermi surfaces. Thus the oscillation period within an RKKY-based model will be determined not by the Fermi wavelength but rather by the detailed topology of the Fermi surface [11,13,14]. In particular, P_{TM} is expected to be related to special wave vectors which span or nest the Fermi surface and which may give rise to large susceptibilities of the electron gas. The magnitude of such wave vectors will usually be smaller than π/λ_F. Thus one could argue that the relatively long oscillation periods found for Cu, Cr, and Ru reflect the special morphology of the Fermi surfaces for each of these metals. Furthermore, in such a model the strength of the magnetic coupling, J_{TM}, would also be determined, in large part, by the detailed Fermi surface topology [5,11,14]. Thus, intuitively, it would seem unlikely that either the oscillation period or the magnetic coupling strength would vary systematically among the transition metals.

In this Letter we show first that oscillatory magnetic exchange coupling via the transition metals is a general phenomenon which we have found for eleven different 3*d*, 4*d*, and 5*d* transition metals sandwiched between Fe, Co, Ni, or Ni alloy magnetic layers. Second, we show that the period of the oscillations in magnetic coupling is approximately the same, $P \simeq 9$–11 Å, for all of these elements with the exception of Cr. Finally, we demonstrate that the magnitude of the coupling strength varies systematically within the transition metals. In particular, the strength of the coupling systematically increases from the 5*d* to 4*d* to 3*d* elements and increases *exponentially* with the number of *d* electrons along the 3*d*, 4*d*, and 5*d* periods.

Oscillatory magnetic exchange coupling through transition metals was first observed in sputtered polycrystalline Fe/Cr, Co/Cr, and Co/Ru multilayers [1]. For these studies the sputter-deposition process has two advantages compared to molecular-beam-epitaxy growth procedures. First, it is possible to prepare large numbers of samples under very similar conditions in highly automated sputter-deposition systems. Second, we find that typical sputter-deposition growth conditions lead to the formation of more complete thin-film layers and consequently better-defined compositional profiles in many cases. Films were prepared for these studies by magnetron sputter deposition in an automated high-vacuum system with a base pressure of $\simeq 2\times 10^{-9}$ Torr. All the structures were grown under similar conditions at $\simeq 40$ °C, in 3.25 mTorr Ar at deposition rates of $\simeq 2$ Å/sec, on chemically etched Si(100) wafers. These studies involved more than 1500 samples. The multilayers are polycrystalline, textured (111), (110), and (0001) for fcc, bcc, and hcp structures, respectively.

Perfect antiferromagnetic coupling of successive magnetic layers in sandwich structures or multilayered structures containing an even number of magnetic layers implies that in zero field the net magnetization is zero. Application of a magnetic field will tend to align the magnetic moments of the individual magnetic layers such that for fields larger than a saturation field H_S these moments are completely aligned [15]. The magnitude of the antiferromagnetic (AF) interlayer magnetic exchange coupling is given by $J_{AF} = H_S M t_F/2\alpha$, where M and t_F are the magnetization and thickness, respectively, of the ferromagnetic layers, and α varies from 1 in simple sandwich structures to 2 as the number of magnetic layers becomes very large [16]. The antiferromagnetic arrangement of the magnetic layers in small magnetic fields has

been confirmed in sputtered polycrystalline Fe/Cr and Co/Ru multilayers [16,17] and in single-crystal Co/Cu and Fe/Cr multilayers [18,19] by neutron-scattering experiments. The Co/Ru system neutron-scattering studies have, in addition, confirmed oscillations in the antiferromagnetic coupling with Ru thickness [17].

First, we consider multilayers where the magnetic layers are comprised of Co. Structures of the form Si(100)/(buffer layer)/[Co(15 Å)/TM]$_N$/(capping layer) were prepared as described above. The number of bilayers, N, was usually 16. Buffer and capping layers of $\simeq$30–50 Å Cr were used in most cases, because Cr does not react with the silicon wafer and forms well-defined flat layers as determined from cross-section transmission electron micrographs. Families of multilayers were prepared using almost all of the 3d, 4d, and 5d transition metals, with the obvious exception of Tc. No structures were prepared using Os or Sc because of their high cost, Mn because of its reactivity, and Zn and Cd because of the possibility of residual contamination of the deposition system.

Typical magnetization versus in-plane magnetic-field hysteresis loops for representative multilayered samples comprised of Co layers separated by eight different elements, V, Nb, Mo, Rh, Ta, W, Re, and Ir, are shown in Fig. 1. Such loops are similar to those found for similarly prepared antiferromagnetically coupled Co/Cr, Co/Ru, and Co/Cu multilayers [1,20]. They thus provide evidence of antiferromagnetic exchange coupling of Co layers separated by a large number of additional transition-metal layers. As can be seen from Fig. 1, the saturation fields, and thus the magnitude of J_{AF}, vary considerably depending on the transition metal. Moreover, just as for earlier studies on Co/TM multilayers, where TM=Cr, Ru, or Cu, the magnetization of structures with slightly thinner or thicker spacer layers saturates in much smaller fields, determined by the coercive field of the magnetic layers, with no evidence for AF coupling. Furthermore, for all of these elements except Nb, Ta, and W (where the antiferromagnetic coupling is very weak), several oscillations in the saturation field are found as the thickness of the spacer layer is varied. Examples of typical data are shown for three Co/TM multilayers in Fig. 2. The saturation field is seen to oscillate with oscillation periods of approximately 9, 11, and 9 Å, for V, Mo, and Rh, respectively. These oscillation periods can be compared with the earlier results of 10 Å in Co/Cu multilayers [2] and 11 Å in Co/Ru multilayers [1]. Remarkably, the oscillation periods are approximately the same in each case irrespective of the transition metal. The only exception is Cr, for which the period is approximately 18–20 Å [1,8]. Note that Cr is unique among these elements in that it displays magnetic character. Bulk Cr orders below 310 K into an unusual linear spin-density-wave (SDW) antiferromagnetic state characterized by an extremely long wavelength of $\simeq$40 Å [21]. In the SDW state the magnetic moments on the Cr alternate in direction from one

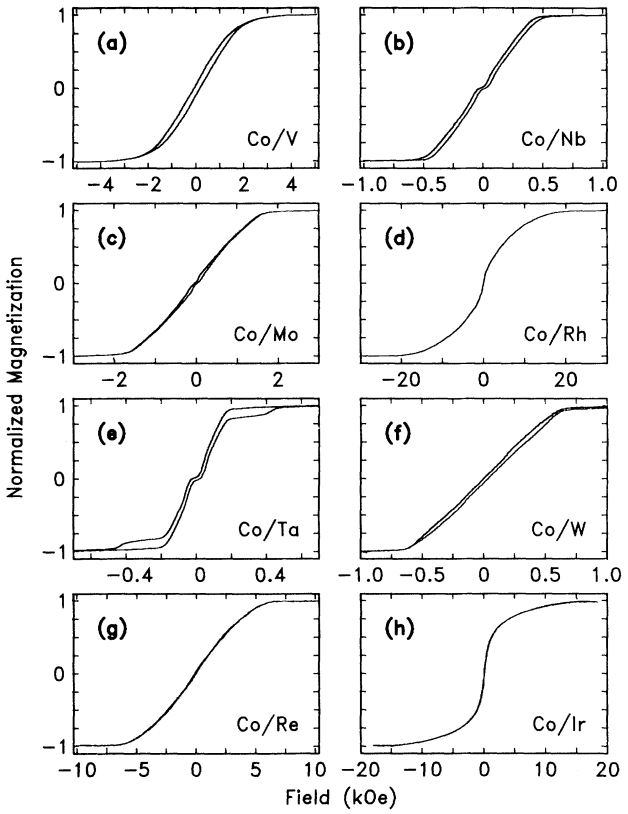

FIG. 1. Magnetization vs in-plane magnetic-field loops for multilayers with spacer-layer thicknesses close to the first antiferromagnetic oscillation. (a) Si/Cr(35 Å)/[Co(16 Å)/V(9 Å)]$_{16}$/Cr(20 Å), (b) Si/Cr(35 Å)/[Co(15 Å)/Nb(6.8 Å)]$_{16}$/Cr(20 Å), (c) Si/Cr(35 Å)/[Co(15 Å)/Mo(5.2 Å)]$_{16}$/Cr(20 Å), (d) Si/Rh(50 Å)/[Co(20 Å)/Rh(7.9 Å)]$_{16}$/Rh(40 Å), (e) Si/Cr(35 Å)/[Co(15 Å)/Ta(7.1 Å)]$_{16}$/Cr(20 Å), (f) Si/Cr(35 Å)/[Co(15 Å)/W(5.3 Å)]$_{16}$/Cr(20 Å), (g) Si/Re(135 Å)/[Co(20 Å)/Re(5.4 Å)]$_{20}$/Re(75 Å), and (h) Si/Ir(110 Å)/[Co(17 Å)/Ir(4.3 Å)]$_{20}$/Ir(11 Å).

Cr site to the next. An oscillation with such a short period has recently been observed in Fe/Cr/Fe(100) wedges grown on perfect Fe whiskers [9]. While oscillations with such short periods would be masked by interface roughness in our structures, oscillations with periods ranging from $\simeq$5 Å to more than 50 Å should be identifiable. No evidence for antiferromagnetic coupling was found for Co/TM multilayers containing Ti, Zr, or Hf or Pd, Pt, Ag, or Au spacer layers [22].

Table I shows a compilation of room-temperature results on Co/TM multilayered structures. The table includes values of (a) the spacer-layer thickness, A_1 (Å), corresponding to the position of the first peak in antiferromagnetic exchange-coupling strength as the spacer-layer thickness is increased; (b) the magnitude of the antiferromagnetic exchange-coupling strength, J_1 (erg/cm^2), at this first peak; (c) the approximate range of spacer-layer thickness of the first antiferromagnetic region, ΔA_1 (Å); and finally (d) the oscillation period, P

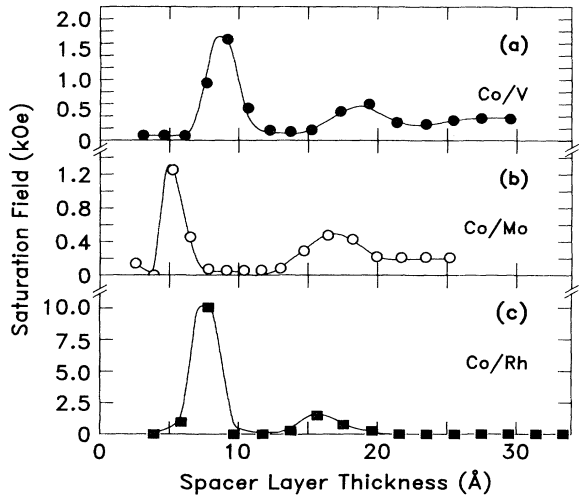

FIG. 2. Dependence of saturation field on spacer-layer thickness for families of Co/V, Co/Mo, and Co/Rh multilayers. Hysteresis loops for one member of each of these families are shown in Figs. 1(a), 1(c), and 1(d), respectively. The saturation field is here defined as the field corresponding to 80% of the saturation moment of the sample averaged over the four quadrants of the hysteresis loop.

(Å). Note that A_1, ΔA_1, and P display no significant temperature dependence, but that J_1 typically increases by $\simeq$20%–40% as the temperature is decreased from 300 to 4.5 K. As can be seen from the table, ΔA_1 is approximately the same for all the elements except for Cr, for which it is about twice as long Thus, as expected, ΔA_1 scales with the oscillation period. This means therefore that for those elements for which the coupling is very weak and only a single AF region is found, ΔA_1 can be used to infer their expected oscillation period. For Nb, Ta, and W, the values of ΔA_1 given in Table I suggest $P_{TM} \simeq 10$ Å very similar to those for the other elements (except Cr). The observed oscillation periods are, as mentioned earlier, much longer than continuum RKKY models would predict. Taking into account the discrete lattice leads to longer oscillation periods [10] and multiperiodicity [11]. In contrast to the oscillation period, the oscillation phase A_1 varies considerably from TM to TM (see Table I). Indeed, A_1 varies widely even for the same TM when sandwiched between different magnetic layers [1,23,24]. One obvious possibility is that intermixing at the interfaces, to a greater or lesser degree, would affect A_1. The oscillation phase is also expected to be affected by the local interaction between the magnetic and spacer-layer atoms and the topology of the Fermi surface.

The magnitude of the interlayer exchange coupling falls off with increasing spacer-layer thickness t, as described by some envelope function. Thus the wide variation in A_1 means we cannot use values of J_1 to directly compare coupling strengths for different TM, since J_1 samples the envelope function at different t corresponding to A_1. In order to compare the intrinsic coupling

TABLE I. Perioic table of A_1 (Å), the spacer-layer thickness corresponding to the position of the first peak in antiferromagnetic exchange-coupling strength as the spacer-layer thickness is increased; J_1 (erg/cm^2), the magnitude of the antiferromagnetic exchange-coupling strength at this first peak; ΔA_1 (Å), the approximate range of spacer-layer thickness of the first antiferromagnetic region; and P (Å), the oscillation period. The most stable crystal structure of the various elements is included for reference, as well as values of the Wigner-Seitz radii [r_{WS} (Å)]. Note that no dependence of the coupling strength on crystal structure nor any correlation with electron density ($\propto r_{WS}^{-3}$) is found. An asterisk indicates that, as discussed in the text for the elements Nb, Ta, and W, only one AF-coupled spacer-layer thickness region was observed, so it was not possible to directly determine P.

Ti ⬡	V ⬢		Cr ⬢		Mn ⬟	Fe ⬢	Co ⬡	Ni ▣	Cu ▣	
No Coupling	9	3	7	7		Ferro-Magnet	Ferro-Magnet	Ferro-Magnet	8	3
	0.1	9	.24	18					0.3	10
2.89	2.62		2.50		2.24	2.48	2.50	2.49	2.56	
Zr ⬡	**Nb** ⬢		**Mo** ⬢		**Tc** ⬡	**Ru** ⬡	**Rh** ▣	**Pd** ▣	**Ag** ▣	
No Coupling	9.5	2.5	5.2	3		3	3	7.9	3	No Coupling	No Coupling
	.02	*	.12	11		5	11	1.6	9		
3.17	2.86		2.72		2.71	2.65	2.69	2.75	2.89	
Hf ⬡	**Ta** ⬢		**W** ⬢		**Re** ⬡	**Os** ⬡	**Ir** ▣	**Pt** ▣	**Au** ▣	
No Coupling	7	2	5.5	3	4.2	3.5		4	3	No Coupling	No Coupling
	.01	*	.03	*	.41	10		1.85	9		
3.13	2.86		2.74		2.74	2.68	2.71	2.77	2.88	

▣ fcc ⬢ bcc
⬡ hcp ⬟ complex cubic

Element	
A_1 (Å)	ΔA_1 (Å)
J_1 (erg/cm^2)	P (Å)
r_{ws} (Å)	

strengths for different TM, it is necessary to make some assumption about the form of the envelope function. In the simplest RKKY model, assuming no structural dependences on t, the envelope function varies as $1/t^2$ for large t [13]. Although exchange coupling via Ru is well described by such a function [23,24], for many other TM the coupling falls off much more rapidly (see Fig. 2 and Refs. [1,8]). Nevertheless, as a first approximation, let us assume that the exchange-coupling strength does vary as $1/t^2$. This allows us to define an equivalent coupling strength for $t=3$ Å as $J_0 \equiv J_1(A_1/3)^2$. Values of J_0 derived from J_1 and A_1 given in Table I are plotted in Fig. 3(a) versus the number of TM valence electrons for various Co/TM multilayers. The figure clearly shows that J_0 increases along each of the 3d, 4d, and 5d periods and along each column from the 5d to 3d metals. Moreover, it also indicates that the effective exchange-coupling strength increases approximately exponentially with the number of d electrons along the 4d and 5d periods. A similar result is also found for Fe/TM, Ni/TM, Ni_{80}-Co_{20}/TM, and $Ni_{80}Fe_{20}$/TM multilayers. However, a less complete data set is available for these structures. In

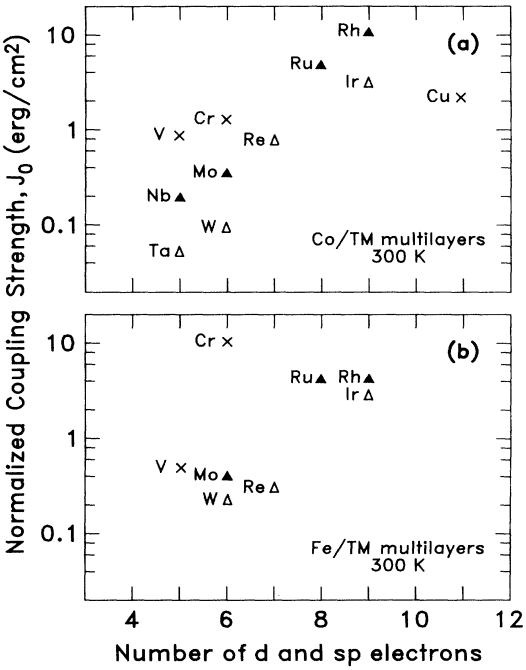

FIG. 3. Dependence of the normalized exchange coupling constant on the 3*d*, 4*d* and 5*d* transition metals in (a) Co/TM and (b) Fe/TM multilayers.

particular, oscillatory AF coupling was found in Fe/TM structures containing V,Cr, Mo,Ru,Rh, and W,Re,Ir, but no coupling was found for structures containing Cu, Nb, or Ta [25]. In general, the Fe/TM structures were of a poorer structural integrity compared to comparable Co/TM structures. However, the dependence of coupling strength on TM is similar for the Fe/TM structures. For the 3*d* elements, J_0 increases from V to Cr, but Cu appears to be anomalous, perhaps due to its filled 3*d* band.

Neither the similarity of the oscillation period nor the systematic variation of coupling strength among the transition metals can be easily understood by examining the Fermi surfaces of the various TM. Moreover, no correlation of coupling strength with the density of states at the Fermi level (total, *d* or *sp* projected densities [26]), occupied bandwidth [26], or such properties as magnetic susceptibility [27] is found. In any case, these data provide a beautiful monitor of the incipient tendency of the various transition metals towards magnetism. In this regard we note an inverse correlation of the exchange-coupling strength with superconductivity [28].

In summary, we have demonstrated that oscillatory exchange coupling is a general phenomenon among the transition metals. Remarkably, we find that for all these metals, with the exception of Cr, the oscillation period is approximately the same, about 10 Å. Moreover, the indirect exchange-coupling strength varies systematically throughout the transition metals, increasing from the 5*d* to 4*d* to 3*d* metals and increasing exponentially along each period. Surprisingly, no dependency of either the oscillation period or the coupling strength on the different crystal structures of the various transition metals was found.

I thank K. P. Roche for technical support.

[1] S. S. P. Parkin *et al.*, Phys. Rev. Lett. **64**, 2304 (1990).
[2] S. S. P. Parkin *et al.*, Phys. Rev. Lett. **66**, 2152 (1991).
[3] Y. Wang *et al.*, Phys. Rev. Lett. **65**, 2732 (1990).
[4] P. M. Levy *et al.*, J. Appl. Phys. **67**, 5914 (1990).
[5] D. M. Edwards *et al.*, Phys. Rev. Lett. **67**, 493 (1991).
[6] W. R. Bennett *et al.*, Phys. Rev. Lett. **65**, 3169 (1990).
[7] A. Cebollada *et al.* (unpublished).
[8] S. Demokritov *et al.*, Europhys. Lett. **15**, 881 (1991).
[9] J. Unguris *et al.*, Phys. Rev. Lett. **67**, 140 (1991).
[10] C. Chappert and J. P. Renard, Europhys. Lett. **15**, 553 (1991); D. M. Deaven *et al.*, Phys. Rev. B **44**, 5977 (1991); R. Coehoorn (unpublished).
[11] P. Bruno and C. Chappert, Phys. Rev. Lett. **67**, 1602 (1991).
[12] K. Moorjani and J. M. D. Coey, *Magnetic Glasses* (Elsevier, New York, 1984).
[13] C. Kittel, in *Solid State Physics*, edited by F. Seitz, D. Turnbull, and H. Ehrenreich (Academic, New York, 1968), Vol. 22, p. 1.
[14] L. M. Roth *et al.*, Phys. Rev. **149**, 519 (1966).
[15] P. Grunberg, in *Topics in Applied Physics: Light Scattering in Solids V*, edited by M. Cardona and G. Guntherodt (Springer-Verlag, Berlin, 1989), Vol. 66, p. 303.
[16] S. S. P. Parkin *et al.*, Appl. Phys. Lett. **58**, 1473 (1991).
[17] Y. Y. Huang *et al.*, J. Magn. Magn. Mater. **99**, L31 (1991).
[18] A. Cebollada *et al.*, Phys. Rev. B **39**, 9726 (1989).
[19] A. Barthelemy *et al.*, J. Appl. Phys. **67**, 5908 (1990).
[20] S. S. P. Parkin *et al.*, Appl. Phys. Lett. **58**, 2710 (1991).
[21] E. Fawcett, Rev. Mod. Phys. **60**, 209 (1988).
[22] We cannot rule out the possibility that structural defects could obscure the presence of any antiferromagnetic interlayer coupling. In particular, subtle defects such as pinholes from one magnetic layer to the next could give rise to large direct exchange coupling which could overwhelm the much smaller indirect coupling.
[23] S. S. P. Parkin and D. Mauri, Phys. Rev. B **44**, 7131 (1991).
[24] S. S. P. Parkin (unpublished).
[25] Note that antiferromagnetic coupling has been observed in molecular-beam-epitaxy grown Fe/Cu/Fe sandwiches with a coupling strength comparable to that which we find in sputtered Co/Cu multilayers [see B. Heinrich *et al.*, Phys. Rev. Lett. **64**, 673 (1990), and Ref. [6]].
[26] D. A. Papaconstantopoulos, *Handbook of the Band Structure of Elemental Solids* (Plenum, New York, 1986).
[27] *CRC Handbook of Chemistry and Physics*, edited by R. C. Weast, M. J. Astle, and W. H. Beyer (CRC, Boca Raton, 1987).
[28] N. F. Berk and J. R. Schrieffer, Phys. Rev. Lett. **17**, 433 (1966).