Theory of Oscillatory Exchange Coupling in Fe/(V,Cr) and Fe/(Cr,Mn)

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Using a first-principles method, we determined the composition and orientation dependence of oscillatory exchange coupling in bcc Fe/V_xCr_{1-x} and $Fe/Cr_{1-y}Mn_y$ magnetic multilayer alloys for $0 < x < 1$ and $0 < y < 0.3$. For the [001] and [110] orientations, the coupling was oscillatory and well described by the superposition of two well-separated RKKY-like components. The composition dependence of the coupling amplitudes and periods shows a rich variety of features. These are discussed and compared with experiment and the predictions of simplified models.

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Because of its intrinsic scientific interest and relevance to giant magnetoresistance, the oscillatory exchange coupling observed in magnetic multilayers [1] is now widely studied [2]. Recently, we showed how the exchange coupling in idealized bcc [001] Fe/V and Fe/Cr multilayers can be determined from first principles [3]. Here we undertake the more difficult task of determining the exchange coupling in related multilayer alloys, namely, bcc [001] and [110] Fe/(V,Cr) and Fe/(Cr,Mn). Earlier studies of multilayers with alloy spacers focused on Fe/Ni_xCu_{1-x}, where the x dependence of the oscillatory periods can be readily determined from their simple Fermi surfaces [4]. The present study investigates transition metal alloy spacers having complex Fermi surfaces.

In an earlier paper [3] we studied exchange coupling in Fe/Cr with paramagnetic (PM) as well as antiferromagnetic (AFM) spacers. Here we limit ourselves to PM spacers, because the AFM character of Cr-rich spacers is normally suppressed by interfacial roughness unless the samples are of high quality, as in Fe/Cr whiskers [5]. We were motivated to study the Fe/(V,Cr) multilayer because experimental results are available (cf. Fig. 2), and because the Fermi surfaces are too complex to be analyzed by topological considerations alone. We also consider $Fe/Cr_{1-y}Mn_y$, for $y < 0.3$. Self-consistent calculations show that multilayers with $y < 0.3$ exhibit the same short-period AFM behavior as Cr, which will be similarly suppressed by interfacial roughness. Thus our use of PM spacers is justified for the compositions we consider. It avoids the need to treat the complex magnetic behavior inherent in idealized or Mn-rich interfaces [6].

Following the approach of Ref. [3], we carry out first-principles multilayer calculations within the framework of the collinear local spin density approximation (LSDA). The zonal integrations were done with an improved tetrahedron method [7] using 600 to 2500 inequivalent points: The integrations were converged to a few μRy per unit cell. We represent the magnetic slabs by 2 Fe monolayers (ML) and the spacers by (V,Cr) or (Cr,Mn) alloys whose thickness ranges from 6 to 32 ML for [001] and from 6 to 24 ML for [110]. For Fe/V_xCr_{1-x}, the interplanar spacing d is interpolated linearly between 3.02 and 2.87 Å, while the planes connecting Fe layers are compressed to conserve the Fe atomic volume. For (Cr,Mn), all atoms lie on a common bcc lattice, with $d = 2.87$ Å.

In substitutional alloys (V,Cr) and (Cr,Mn), the parameters that dominate the alloy scattering (bandwidth difference and center of gravity difference) are small. Disorder changes the real part of the effective alloy potential, altering the shape of the bands and the Fermi surface. It also adds an imaginary component to the potential, which, for weak scattering, broadens the energy levels and gives them finite lifetimes [8]. To test the effect of disorder quantitatively, we compared bulk $V_{0.5}Cr_{0.5}$ and $Mn_{0.3}Cr_{0.7}$ within the coherent potential approximation (CPA), with that given by the virtual crystal approximation (VCA). The average of the effective CPA V and Cr (or Cr and Mn) potentials differed minimally (<1 mRy) from the corresponding average of the bulk potentials (VCA). Moreover, we found negligible differences in the Fermi surfaces of an ordered compound of V and Cr (CsCl structure) and CPA and VCA versions of this material. Apart from phase shifts, we also found that exchange coupling was very similar in a $Fe/V_{0.5}Cr_{0.5}$ multilayer with the spacer treated as a virtual crystal and as an ordered CsCl compound. Finally, broadening of the levels of $V_{0.5}Cr_{0.5}$ and $Mn0.3Cr_{0.7}$, as calculated within the CPA, was found to be very small compared with a typical effective radius of curvature of the Fermi surface.

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Based on these findings, we feel justified in using the VCA in all our calculations. Within the atomic sphere approximation (ASA), a trial charge density can be completely determined by taking the compositionally weighted average of the lowest three energy moments and nuclear charges Z of the respective bulk materials. This form of the VCA is especially convenient because (V,Cr) and (Cr,Mn) may be regarded as continuous functions of Z, which varies from 23 for pure V, to 24.3 for $Cr_{0.7}Mn_{0.3}$.

In Fig. 1 we display the calculated coupling strengths for [001] and [110] as functions of spacer thickness for selected compositions. The coupling strength, given by the total energy difference [9] $E_x = E[Fe_2^{\uparrow}B_mFe_2^{\downarrow}B_m] - E[Fe_2^{\uparrow}B_mFe_2^{\downarrow}B_m]$—with $B(Z)$ regarded as an "element" with $Z = 24 - x$ for V_xCr_{1-x} and $Z = 24 + y$ for $Cr_{1-y}Mn_y$—is seen to be oscillatory with multiple periods. Accordingly, we attempt to describe the coupling by fitting the data of Fig. 1 for each orientation and composition to the sum of two RKKY-like terms:

$$E_x(m) = \sum_{j=l,s} A_j m^{-p_j} \cos\left(\frac{2\pi m}{T_j} + \phi_j\right),$$

where the long(er) and short(er) periods are denoted by l and s, and the amplitudes, periods, and phases by T, A, and ϕ. If Fermi surface nesting occurs at isolated points, the envelope power $p = 2$. However, for nesting over finite regions, as in Cr and Cr-rich alloys, p can be as small as unity. Fitting the eight undermined parameters A_j, T_j, ϕ_j, and p_j $(j = l, s)$ to the data in Fig. 1, and excluding $m < 6$ to avoid the preasymptotic range, p turns out to be approximately 2 for all cases except the [001] orientation for $Z \geq 23.9$, where both p are approximately 1.25. To simplify our presentation, we refitted the data, constraining p_l and p_s to be 1.25 for the [001] orientation at $Z \geq 23.9$, and 2 for all other cases. This constraint did not increase the fitting errors significantly. The resulting amplitudes and periods are shown in Fig. 2. There are several points of interest:

(a) Our two-period RKKY-like model fits the data quite well except for $Z = 23.9$ along [110], where more than two prominent components may be present. Indeed, a Fourier transform of the data shows a spread of the short-period component over a range of about 2 ML, while the long period is clearly resolved. Thus, the fitted short period represents an average of a cluster of short-period components. We were not able to improve the fit here or elsewhere using three periods. For pure V and Cr, the periods we find are close to some of these predicted by Stiles's Fermi surface analysis [10]. Stiles also found many other periods, but these are apparently too weak to give rise to significant coupling.

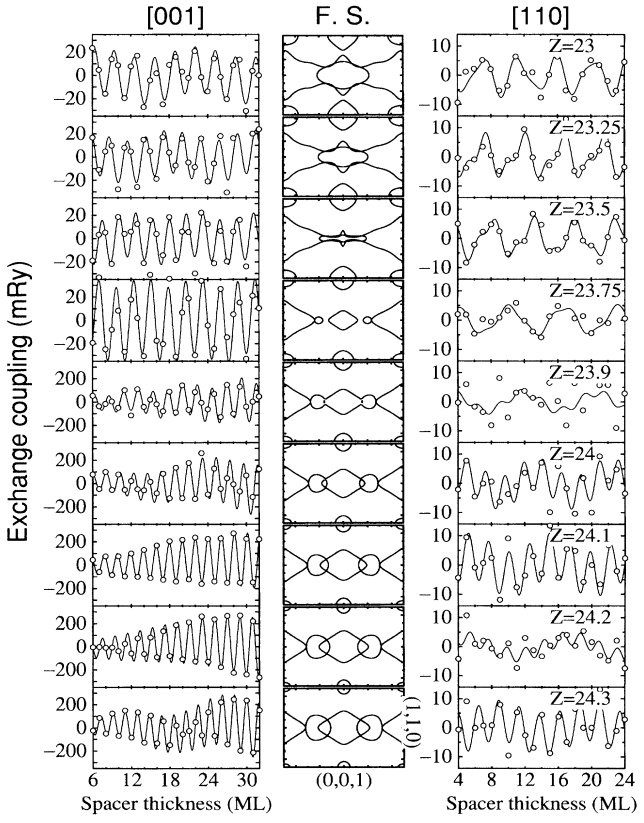

FIG. 1. Open circles, left and right panels: calculated $E_x \times m^2$ vs m for various alloy compositions for [001] and [110] orientations. Fits are denoted by solid lines. The central panel shows the corresponding Fermi surfaces profiles.

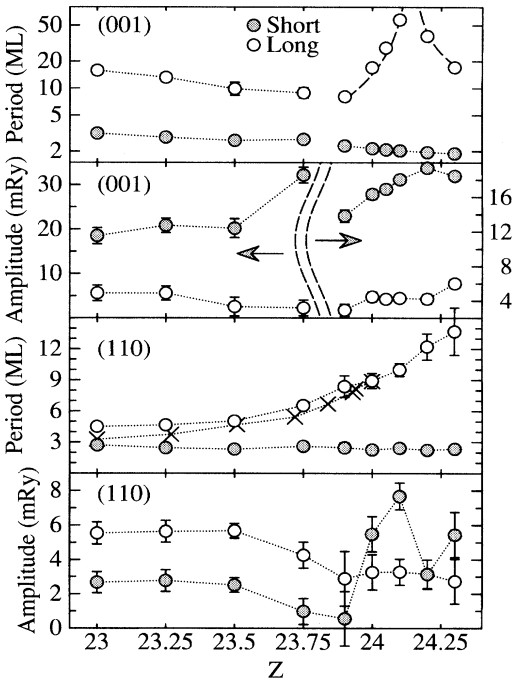

FIG. 2. Amplitudes and periods vs Z for [001] and [110] orientations. The break in the [001] amplitudes corresponds to a change in the envelope power p. Crosses denote experimental points. The error bars mark 1/4 of a standard deviation, which is also approximately the range of values obtained by varying the range of m used for the fits.

(b) There are abrupt changes in the [001] coupling near $Z = 23.9$: E_x increases by a factor of ~ 4 between $Z = 23.75$ and $Z = 23.9$ (cf. Fig. 1), and p changes from 2 to 1.25, as noted previously. These effects are associated with the onset of large, approximately polygonal Fermi surfaces centered at Γ and H, for which there is nesting over large regions of $k_\parallel$. As Z varies, the relative size of their faces changes. They reach the same size at $Z = 24.15$, where the short period becomes exactly 2 ML. The short-period coupling strength is greatest near $Z = 24.15$, presumably because the nesting vectors have their maximal cross-sectional area there.

(c) Moreover, the long period exhibits a remarkable divergence at $Z = 24.15$. For $Z > 23.9$, T_l is apparently related to T_s by $T_l = T_s/(2 - T_s)$ (dashed lines). In Ref. [11], it was noted that the coupling is described by a distorted wave form; the second harmonic can appear as an effective long-period oscillation with $T_l = T_s/(2 - T_s)$. For $Z < 23.9$, this relationship between shorter and longer periods breaks down. Here the two periods arise from different nesting features.

(d) For the [110] orientation, the theoretical Z dependence of T_l agrees very well with the measured behavior [12], bearing in mind that the experimental periods are only approximate, having been extracted from data for only two peaks. The short period T_s was not observed because it is predicted to be weaker than T_s and, in addition, likely to be suppressed by interfacial roughness.

(e) The theoretical composition dependence of the coupling strength is generally consistent with measurements by Takanashi *et al.* [13]. However, these comparisons are not conclusive because their interfaces did not have well-defined orientations. According to our theory, the Z dependence of T_s is slow and weakly orientation dependent, while T_l should show a strong orientation dependence. Experiments using well-oriented interfaces are in progress to test these theoretical predictions.

(f) For $Z \leq 23.75$ and both orientations, the amplitude remains flat or decreases with Z. This behavior is opposite to predictions based on simplified theories [14,15].

(g) Our calculated coupling strength for [001] Fe/Cr is approximately 10 times larger than the best measured values. We noted earlier [3,16] that this discrepancy can be attributed to interfacial mixing of Fe and Cr atoms. To estimate how much this coupling is quenched, we calculated E_x for an [001] Fe/Cr multilayer, in which the interfacial layer is composed equally of Fe and Cr (checkerboard geometry). E_x exhibited oscillatory coupling with periods essentially the same as that for the ideal interface. However, the short- and long-period amplitudes dropped by factors of 6.1 and 5.8, respectively, demonstrating that interfacial intermixing can significantly reduce the coupling strength. We cannot expect to obtain perfect agreement between theory and experiment because our knowledge of atomic-scale intermixing at real interfaces is still rudimentary [17].

(h) In relatively high quality [001] Fe/Cr samples, short as well as long periods are observed, and the coupling does not oscillate about zero for thin spacers, but possesses a (AFM) bias [18]. It has been suggested—but not demonstrated conclusively—that this bias is a manifestation of superexchange [19] which lies outside the LSDA. Indeed, our own LSDA calculations and those of others [20] do not show this bias. Further experiments should be carried out to ascertain whether the observed AFM bias is an intrinsic effect, especially in light of its observed temperature dependence, which remains unexplained by any theory.

In conclusion, we have predicted the periods most likely to be observed, as well as their composition and orientation dependence. We predict a wealth of new effects in [001] and [110] Fe/(V,Cr) and Fe/(Cr,Mn); for example, the long period in [001] $Fe/Cr_{1-y}Mn_y$ tends to ∞ near $y = 0.15$. We also show that interfacial intermixing can strongly quench E_x, and thus explain why first-principles calculations of idealized interfaces tend to overestimate the strength of the observed coupling. Our results should prove helpful in testing simplified theories, guiding future experiments, and designing optimized materials.

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