# Simplified First-Principles Approach to Exchange Coupling in Magnetic Multilayers

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We determined oscillatory periods, phases, and coupling strengths for [001] Fe/V and Fe/Cr, using a highly accurate, non-self-consistent first-principles method. The periods obtained for Fe/Cr are consistent with experiment both for paramagnetic and antiferromagnetic Cr. These results clearly show how the Cr antiferromagnetism affects the exchange coupling. For [001] Fe/V, we predict short and long periods that have not yet been observed, but provide tests for the theory. We also discuss the merits of our computational approach.

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The recent discovery [1] that the exchange coupling $E_x$ between successive magnetic slabs in multilayers such as Fe/Cr and Co/Cu is an oscillatory function of the spacer thickness is of great scientific interest [2]. Further interest in this subject arises from the related discovery of giant magnetoresistance in antiferromagnetically coupled multilayers [3] and possible applications to magnetic recording technology. Although oscillatory coupling has been observed in a wide variety of multilayer systems [4], it has proved difficult to predict the oscillatory periods in multilayers not having simple Fermi surfaces [5,6].

Here we describe a simplified first-principles method for determining $E_x$ for arbitrary magnetic multilayers. Our approach retains the precision and relatively rigorous foundation of the local spin density approximation (LSDA) [6], while offering the efficiency of empirical tight-binding calculations. In contrast to model calculations [5,7], we are able to determine oscillatory periods, phases, and coupling strengths of realistic multilayers having complex Fermi surfaces. The present approach is considerably more efficient than fully self-consistent calculations since it avoids the extremely slow convergence associated with instabilities (charge sloshing) inherent in large unit cells. Moreover, we are able to achieve $\mu$Ry accuracy, which is crucial in the present application.

The first key ingredient in our approach is the use of a non-self-consistent procedure which exploits the fact that self-consistent multilayer charge density deviates significantly from bulk behavior only close to the interface. We approximate the self-consistent density by a suitably chosen "trial" density $n_{\rm in}$ which is exact well away from the interface, and make use of the variational principle which states that errors in the total energy are second order in deviations from the self-consistent density $n_0$. This simplification makes feasible calculations that would otherwise be computationally prohibitive.

Although there is considerable freedom in the choice of the trial density $n_{\rm in}$, its construction must be done with care if one hopes to resolve very small total energy differences between ferromagnetic (FM) and antiferromagnetic (AFM) alignments. Clearly the results are acceptable only if they do not depend significantly on the choice of $n_{\rm in}$. When the coupling is weak, it may be necessary to carry out self-consistent calculations for multilayers with thin spacers to check the validity and improve the accuracy of trial densities. This represents one distinctive feature of our approach. The validity of approximations we make can be tested against the complete theory, and increasingly accurate refinements can be made as needed.

One simple way to construct a trial density for a multilayer is to make self-consistent calculations of the separate elements in their bulk form (e.g., bcc Fe and bcc V), and then "cut and paste" the resulting bulk crystal charge densities into the multilayer environment. This construction is readily accomplished if crystal charge densities are represented by sums of atom-centered densities. Here we employ the atomic spheres approximation (ASA), which represents a crystal charge density by a superposition of overlapping, spherical, and atom-centered densities that fill space [8]. Moreover, the exact potential is approximated by its spherical average inside each sphere. We expect the ASA to introduce only small errors: the dominant errors arising from the neglect of nonspherical contributions in the double-counting terms cancel almost exactly when calculating total energy differences between FM and AFM multilayer alignments.

The second key ingredient is the use of the Harris-Foulkes (HF) energy functional [9]. It is identical to the usual Hohenberg-Kohn (HK) functional, except that double-counting terms are evaluated at the input density $n_{\rm in}$ rather than at the output density $n_{\rm out}$. Because of the large dielectric response of metallic multilayers with thick spacers, small errors in the input potential are greatly amplified, generating large errors in the output potential. The error terms of the HF functional are of order $(n_{\rm in}-n_0)(n_{\rm out}-n_0)$, and considerably smaller than the corresponding $(n_{\rm out}-n_0)^2$ appropriate to the HK.

For these calculations the basis included orbitals to

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$l = 2$, the "combined correction" term was included [8], and the Barth-Hedin exchange functional was employed [10]. Integrations over the Brillouin zone are made with the linear tetrahedron method augmented with Blöchl weights [11], using a mesh of 48×48×4 divisions in the unit cell. Extensive tests showed this mesh was sufficient to converge $E_x$ (calculated as $E_{\mathrm{AFM}} - E_{\mathrm{FM}}$) to within an rms uncertainty of about 2 $\mu$Ry.

Let us first consider bcc [001] Fe/Cr multilayers whose Cr spacers are paramagnetic (PM). We assemble a multilayer trial density using the simple construction mentioned above, placing charge densities obtained from self-consistent calculations of elemental FM Fe and PM Cr at the appropriate positions in the Fe/Cr multilayer. Normally, the input potential $V_{\mathrm{in}}$ would be derived from the input density, i.e., $V_{\mathrm{in}}=V[n_{\mathrm{in}}]$. We exploit the fact that we are free to construct a trial potential $V_{\mathrm{in}}$ independently of $n_{\mathrm{in}}$ to compensate for the dominant error in our construction, namely, that the Fermi levels of bulk Fe and Cr are misaligned. In a self-consistent calculation, this misalignment would be rectified by charge shifting across the Fe/Cr interface, producing interfacial dipoles. Guided by examinations of our Fe/Cr self-consistent multilayer potentials, we compensate for these missing dipoles by inserting an additional step $\Delta V$ into $V[n_{\mathrm{in}}]$ so as to raise the potential in the Fe region relative to that in the Cr. The magnitude of $\Delta V$ is determined variationally by making the total energy stationary. We find that $\Delta V$ depends weakly on spacer thickness and is essentially independent of the magnetic coupling (FM or AFM). As expected, $\Delta V$ turns out to be nearly equal to the Fermi level mismatch between bulk Fe and Cr.

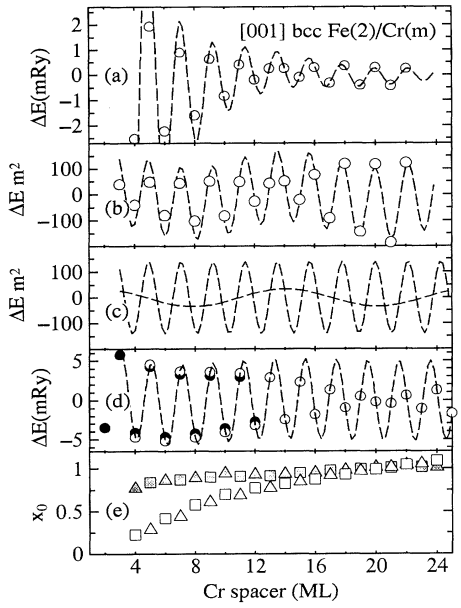


FIG. 1. Exchange coupling $E_x$ in $Fe_2Cr_m$. Circles show calculated $E_x$ vs $m$, using PM Cr as a trial density. Dashed line is a least-squares fit to the asymptotic RKKY form. (b) Same as in (a) except that $E_x m^2$ is shown. (c) Decomposition of fit into short and long periods. (d) $E_x$ using a SDW of Cr as a trial density. Dark circles show results of fully self-consistent calculations. (e) Variationally determined amplitude $x_0^m$ of Eq. (3) for FM (squares) and AFM (triangles) coupling; $x_0$=1 corresponds to a maximum moment of $0.68\mu_B$. Dark symbols correspond to the commensurate SDW, light symbols to the incommensurate.

Employing this simple prescription for multilayers of $Fe_2Cr_m^{PM}$, we carried out a series of total energy calculations for $m = 3 \ldots 22$. For each geometry, all Fe and Cr atoms were located on a common bcc lattice with a lattice constant of $5.42a_0$. The exchange coupling shown in Fig. 1 was obtained as half the total energy difference $E[\mathrm{Fe}_2^{\uparrow}\mathrm{Cr}_m\mathrm{Fe}_2^{\downarrow}\mathrm{Cr}_m] - E[\mathrm{Fe}_2^{\uparrow}\mathrm{Cr}_m\mathrm{Fe}_2^{\uparrow}\mathrm{Cr}_m]$. In the upper panel of Fig. 1, the dots denote the calculated points, and the dashed line a least-squares fit to the asymptotic RKKY form

$$E_x = \sum_{j=1}^{2} A_j \sin(q_j m + \phi_j)/(m+1)^2, \qquad (1)$$

obtained using Prony's method [12]. [For numerical reasons, the fit was actually performed for $E_x(m+1)^2$.] To avoid the preasymptotic range, the points fitted were limited to $6 \le m \le 22$. In this way we determined the two amplitudes $A_j$, the two phases $\phi_j$, and the two periods $T_j = 2\pi/q_j$. These are listed in Table I.

To emphasize the sinusoidal nature of the calculated coupling energies and the quality of fit for large $m$, we display $E_x m^2$ versus spacer thickness in the second panel of Fig. 1. It is evident that two oscillatory components are indeed sufficient to provide a good fit to the data. We determined the two periods to be 2.15±0.01 ML and 12.3±0.2 ML (ML denotes monolayer), or about 3.1 Å and 17.7 Å, respectively. (The error bars were estimated by noting the changes incurred in the calculated periods when data points were added or subtracted.)

The short period is consistent with the well-known nesting vector that gives rise to antiferromagnetism in

TABLE I. Calculated amplitudes $A$ (in mRy), periods $T = 2\pi/q$ (in ML), and phases $\phi$ (in rad) for bcc [001] Fe/Cr and Fe/V, corresponding to Eqs. (1) (PM) and (4) (AFM). Values are quoted for a single formula unit (two interfaces). Results are given for the step potential $\Delta V$ (in mRy) set to 0 and to its variationally determined value. Subscripts $s$ and $l$ denote short and long period components, respectively.

| Spacer | $\Delta V$ | $T_s$ | $T_l$ | $A_s$ | $A_l$ | $\phi_s$ | $\phi_l$ |
|---|---|---|---|---|---|---|---|
| $Cr^{PM}$ | 73 | 2.15 | 12.3 | 139 | 34 | 2.87 | −2.48 |
| $Cr^{PM}$ | 0 | 2.20 | 13.5 | 96 | 18 | 2.52 | −0.27 |
| $Cr^{AFM}$ | 73 | 2.05 | 14.4 | −5.1 | 42 | −1.53 | −1.91 |
| $V^{PM}$ | 20 | 3.08 | 11.0 | 23 | 8.6 | 2.72 | 1.17 |
| $V^{PM}$ | 0 | 3.13 | 11.2 | 26 | 7.6 | 1.47 | 2.92 |

Cr. The long period we obtain for [001] Fe/Cr is strikingly close to the value observed in [001] Fe/Cr whisker experiments [13] for Cr in the PM state, as well as to the value of 18 Å observed in [110] Fe/Cr [4]. We believe that Parkin's Cr spacers were PM since the antiferromagnetism of Cr is undoubtedly suppressed by interfacial roughness in his sputter-deposited samples.

In Fig. 2 and Table I we show the results of similar studies for [001] bcc Fe/V. Note that the coupling strengths in Fe/V are only about 1/4 as large as in Fe/Cr. We can again resolve $E_x$ into two oscillatory components, obtaining periods of 4.7 Å and 16.6 Å, respectively. Since to our knowledge Fe/V [100] has not been studied experimentally, these results provide a clear-cut test of our theory.

We also investigated [001] bcc $Fe_2Cr_m^{AFM}$. Since the LSDA incorrectly predicts Cr to be PM at the observed lattice spacing, special measures had to be taken to construct a suitable trial density. By performing self-consistent calculations at larger lattice constants $a$, we found that bulk Cr becomes AFM with a moment of $0.68\mu_B$ at $a$=5.47$a_0$ (about 0.5% larger than the observed spacing). We then assumed that the "spin density" for AFM Cr could be represented by the difference between AFM Cr and PM Cr charge densities at $5.47a_0$. After symmetrizing this difference so that $\Delta n^+ = -\Delta n^-$, we added this "spin density" in the form of an array of atom-centered spin densities to the PM Cr density. In this way, the net density in $Fe_2Cr_m$ became

$$n(\mathbf{r}) = \sum_{j=1}^{m} n_0(\mathbf{r} - \mathbf{R}_j) + x_j^m \Delta n^+(\mathbf{r} - \mathbf{R}_j). \quad (2)$$

For each $m$, we performed two calculations, choosing the $x_j$ to represent first a *commensurate* and then an *incommensurate* spin density wave (SDW) with exactly one phase slip:

$$x_j^m = \begin{cases} x_0^m \cos(\pi j) & \text{(commensurate)}, \\ x_0^m \cos\left[\pi j \left(1 - \frac{1}{m+1}\right)\right] & \text{(incommensurate)}. \end{cases} \quad (3)$$

In both cases, we determined $x_0^m$ variationally by minimizing the total energy. Since adjacent spins at the Fe/Cr interface are assumed to be aligned antiparallel, introducing phase slip in the second expression causes alternate Fe slabs to spin flip. Thus the two forms correspond to FM and AFM coupling. The forms we chose for the $x_j^m$ were suggested by self-consistent calculations, which we had carried out for $m = 2 \ldots 12$.

Figures 1(d) and 1(e) show the calculated $E_x$ vs $m$ and the variationally determined $x_0^m$, respectively. Figure 1(d) shows a preference for the *commensurate* form (which alternates between FM and AFM coupling with each additional layer) until $m = 21$, at which point a phase slip is favored. This is just what is observed for the coupling on Fe whiskers [14]. $E_x$ was fitted to a high precision by

$$E_x = A_1 \sin(q_1 m + \phi_1) + \frac{A_2}{(m+1)^2} \sin(q_2 m + \phi_2). \quad (4)$$

The six fitting parameters are listed in Table I. Note that the first term does *not* have the $1/m^2$ envelope characteristic of RKKY, although the second one does. Comparisons with the coupling strengths of Fig. 1(a) show that the SDW overwhelms the RKKY interaction, though the long RKKY period can still be resolved. We also found that the AFM coupling is much stronger in the multilayer than in bulk Cr. (The latter is very nearly zero according to the LSDA for this spacing.) We attribute this result to the strong tendency of interfacial Cr to align antiparallel relative to its Fe neighbors [6]. This tendency also explains why in the whisker experiments the AFM behavior persists well above the Néel temperature. Note that the observed incommensurate period $2\pi/q_1$ in the whisker experiments is 2.1 ML, while the calculated period is 2.05 ML. We attribute this discrepancy to small errors in the theoretical AFM Cr Fermi surface.

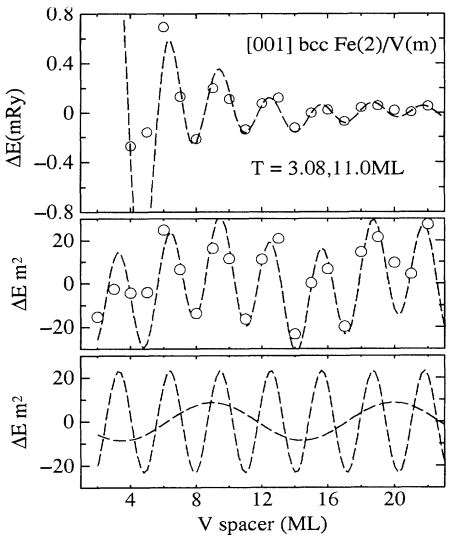


FIG. 2. Exchange coupling $E_x$ in $Fe_2V_m$, analogous to the first three panels of Fig. 1.

In summary, we have described a powerful first-principles method for calculating oscillatory exchange coupling in magnetic multilayers such as Fe/V and Fe/Cr having transition metal spacers with complex Fermi surfaces. Our approach offers the efficiency and flexibility of a tight-binding theory without sacrificing the precision inherent in first-principles methods. Moreover, we can use the input density to do calculations not accessible to a fully self-consistent approach. One important example shown here is the comparison of $E_x$ in Fe/Cr for paramagnetic and antiferromagnetic Cr.

New results derived directly from a local spin density functional theory include (i) a calculation of both a long and a short period in [001] Fe/Cr; (ii) the prediction of a long and a short period in [001] Fe/V; (iii) the comparison of oscillatory coupling in Fe/Cr for PM and AFM spacers; and (iv) the demonstration that oscillatory periods are less sensitive to interfacial potentials than coupling strengths (amplitudes) or phases. Since different degrees of interfacial roughness would give rise to different degrees of interfacial charge transfer and different interfacial potentials, our results suggest that periods are considerably less sensitive to surface conditions than coupling strengths or phases. Finally, we can rule out the need for superexchange [15] terms in describing the oscillatory coupling in Fe/Cr multilayers since we can obtain an excellent description of the experimental results without the inclusion of these terms.

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