Competing Exchange Interactions in Magnetic Multilayers

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We have studied alloying of the nonmagnetic spacer layer with a magnetic material as a method of tuning the interlayer coupling in magnetic multilayers. We have specifically studied the Fe/V(100) system by alloying the spacer V with various amounts of Fe. For some Fe concentrations in the spacer, it is possible to create a competition between antiferromagnetic Ruderman-Kittel-Kasuya-Yoshida exchange and direct ferromagnetic exchange coupling. The exchange coupling and transport properties for a large span of systems with different spacer concentrations and thicknesses were calculated and measured experimentally and good agreement between observations and theory was observed. A reduction in magnetoresistance of about 50% was observed close to the switchover from antiferromagnetic to ferromagnetic coupling.

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Research on multilayers of alternating magnetic and nonmagnetic layers has attracted great interest over the past years because of the successful application of these materials as ultrasensitive hard disk reading heads and magnetic sensors [1–3]. Two important properties of these materials, the oscillating interlayer exchange coupling (IEC) [4] and the giant magnetoresistance (GMR) [5], i.e., a drastic modification in the resistance as a function of an applied external magnetic field, greatly influence the function of these multilayers as magnetic sensors. The development of ways of tailoring and controlling these properties is a crucial step towards the realization of more sensitive magnetic sensors and higher density information storage media. For sensor applications, it is desirable to have a weak antiferromagnetic (AFM) coupling between the magnetic layers.

The mechanism behind the IEC has been identified to be the RKKY coupling [6–8], which is governed by the nesting features of the Fermi surface of the spacer layer material. Methods to tune the strength and sign of the IEC include alloying the spacer with nonmagnetic elements [9–11] or using hydrogen [12]. However, an alternative method can be explored, namely, to introduce an exchange mechanism that competes with the RKKY coupling. The simplest way to achieve this is to alloy the spacer material with a magnetic element and thereby provide a channel of direct metallic exchange. The major advantage over alloying with nonmagnetic elements, where only a change in the Fermi surface topology is expected, is that one can approach gradually a magnetic instability of the spacer layer. As is shown here, this opens up a new possibility of controlling the interlayer exchange coupling. We have carried out our studies using both *ab initio* electronic structure calculations and experiments. We have used the Fe/V multilayer system, with a varying amount of Fe in the V layers, as a model system. The calculations of interlayer exchange interactions have been supplemented by theoretical calculations of the transport properties of these multilayers.

The interlayer exchange coupling (IEC) is defined as the difference in total energy between AFM and ferromagnetically (FM) coupled Fe layers $J = E_{AFM} - E_{FM}$. Our calculations were performed by means of the spin-polarized interface Green's function technique, based on the linear muffin-tin orbitals method within the tight-binding, frozen core and atomic sphere approximations. The method was developed by Skriver and Rosengaard [13]. The alloys were treated within the coherent potential approximation [14–16]. Great care was taken to converge all calculations both in total energy and k-space sampling. We found that 528 k points in the irreducible part of the Brillouin zone (BZ) was sufficient to obtain convergence in all considered cases. The crystal structure was assumed to be bcc with the lattice constant of bulk vanadium.

The transport properties were calculated in a current-perpendicular-to-plane (CPP) geometry, by performing self-consistent calculations using the principal layer technique with 2 atoms per principal layer for a tight-binding Hamiltonian, followed by transport calculations using the Kubo-Landauer approach [17,18]. For the transport calculations of an ideal system a k-point mesh of 10 000 points in the full two-dimensional lateral BZ was used. The specular ($k_{\|}$-conserving) and diffusive ($k_{\|}$-nonconserving) decomposition of the conductance is performed according to the procedure of Ref. [19]. For the disordered system, a (5 × 5) lateral supercell method [17] was used with random occupation of supercell lattice sites by atoms A and B corresponding to the layerwise alloy composition $A_{1-x}B_x$. The conductance was averaged over 3 different supercell alloy configurations, and the maximum variation of conductance between configurations for any spin channel was found to be less than 3%. A k-point mesh corresponding to 6400 k-points in the (1×1) original surface BZ was used.

In our experiments, two series of multilayer films were prepared: $Fe_3/[Fe_xV_{1-x}]_{13}$, $x = 0.00, 0.07, 0.11, 0.19, 0.30$, denoted set 1, and $Fe_3/[Fe_{0.11}V_{0.89}]_n$, $n = 11, 13, 15$ ML, denoted set 2. The two sets represent samples of constant thickness of the Fe_xV_{1-x} layer (set 1) and constant Fe concentration in the vanadium interlayers (set 2). The constant vanadium thickness in set 1 was chosen to be 13 monolayers since the Fe layers of "pure" Fe/V superlattices have the strongest AFM coupling at this thickness of V [20]. All samples were grown on polished MgO(001) $10 \times 10 \times 0.5$ mm^3 single crystal substrates by dc magnetron sputtering from separate Fe (99.95% pure) and V (99.95% pure) targets arranged in a cluster geometry. The growth temperature was 330 °C which is an estimated optimal value obtained from temperature optimization tests performed on Fe/V superlattices [21].

The magnetic measurements were performed in a superconducting quantum interference device magnetometer, Quantum Design MPMSXL at 10 K. All measurements of magnetization (M) vs field (H) were made with the field aligned in plane along the [100] or [110] directions of the Fe/V(Fe)(001) superlattices. The values of the magnetization in Fig. 1 are related to the volume of one Fe/V repetition in the superlattice since it is not straightforward to extract the Fe layer contribution when the spacer is alloyed.

Our samples are almost isotropic, except for the sample with the highest Fe concentration (30%) in the V interlayers which acts almost like a thin film of a homogeneous ferromagnet and shows some in-plane magnetocrystalline anisotropy. As seen in Fig. 1(a), the samples in set 1 show AFM coupling between the Fe layers for iron contents of 0% and 7% in the V interlayers. At higher iron concentrations only weak or FM interlayer interaction occurs (see inset). Complementary magnetoresistance measurements on the sample $Fe_3/[Fe_{0.07}V_{0.93}]_{13}$ showed GMR behavior verifying the AFM character of the interlayer interaction. Turning to the behavior of the samples of set 2, it is seen from Fig. 1(b) that only the sample with V(Fe) interlayer thickness of 15 ML shows AFM coupling between the Fe layers. The results for the two samples with V(Fe) layer thicknesses of 11 and 13 ML indicate weak or FM interaction.

The IEC was calculated for $Fe_3/[Fe_xV_{1-x}]_n$ for the spacer thickness range of 9–17 ML and a concentration range $x = 0$–0.3 in steps of 0.02. Our results for atomically sharp interfaces are shown in Fig. 2 for two choices of spacer layer thickness. The most important information from this figure is that at a concentration of ~20% the

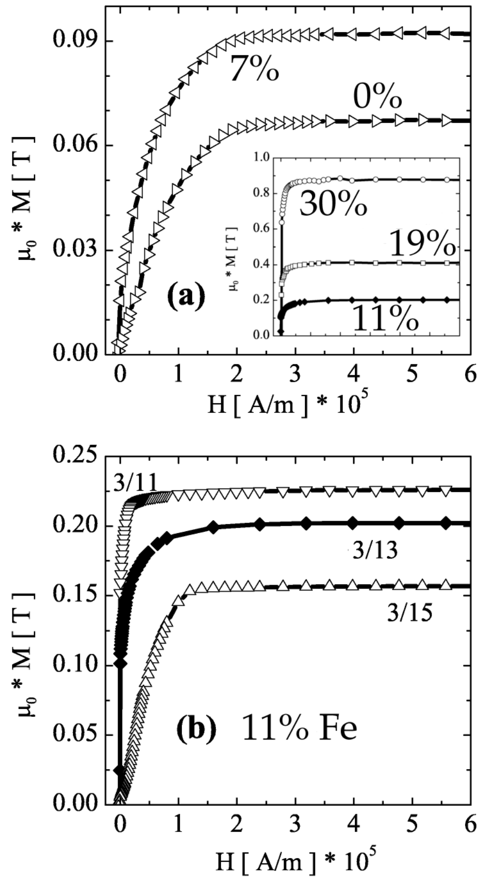

FIG. 1. Field dependence of the magnetization for (a) $Fe_3/[Fe_xV_{1-x}]_{13}$ multilayers with different alloying concentrations in the spacer, (b) $Fe_3/[Fe_{0.11}V_{0.89}]_n$ multilayers with different thicknesses of the spacer. The amplitudes for the AFM coupled systems are Fe_3V_{13}(0%) 0.011 mJ/m^2, Fe_3V_{13}(7%) 0.015 mJ/m^2, and Fe_3V_{15}(11%) 0.013 mJ/m^2, but these values must be seen as a crude estimation due to difficulties to estimate the extent of the magnetic region.

IEC becomes very strong and switches from AFM to FM. Alloying of the spacer layer normally modifies the IEC due to a change of the Fermi surface [7,9,11,22,23]. The transition in Fig. 2 is caused by a different mechanism, since it is due to the fact that at ~20% concentration of Fe in the V layer the spacer layer itself becomes ferromagnetic and the channel for direct exchange opens up. It is of course the Fe atoms in the spacer layer that for these concentrations develop the magnetic moments. As an example we mention that for the $Fe_3/[Fe_xV_{1-x}]_{11}$ multilayer the Fe atoms in the center of the spacer layer have a magnetic moment that is smaller than 0.03 μ_B/atom when $x \leq 0.20$ (in the AFM regime) and that for larger concentrations (i.e., when the IEC becomes FM) the moment increases dramatically to be over 1 μ_B/atom when $x = 0.24$. This rapid onset of ferromagnetism as function of increasing Fe concentration is consistent with experiences from bulk Fe-V alloys, that are paramagnetic for Fe concentrations below 20–30%, and ferromagnetic for larger concentrations [24].

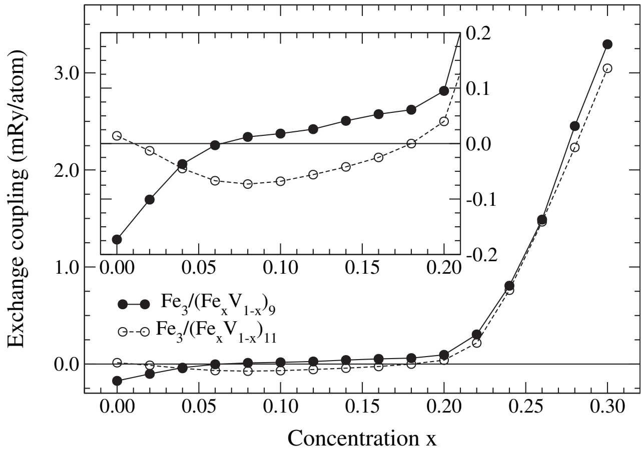

FIG. 2. Calculated interlayer exchange coupling for two Fe/V multilayers with atomically sharp interfaces as a function of Fe concentration in the spacer layers. The inset shows a blowup of the concentration region with less than 20% Fe.

In Fig. 3 we collect all our calculated and measured results of the dependence of the IEC upon spacer layer thickness and alloying concentration of the spacer layer. We have included interface alloying and interface roughness in the calculations by using the method outlined in Ref. [25]. Interface alloying and roughness are known to reduce the coupling strength as compared to the case with

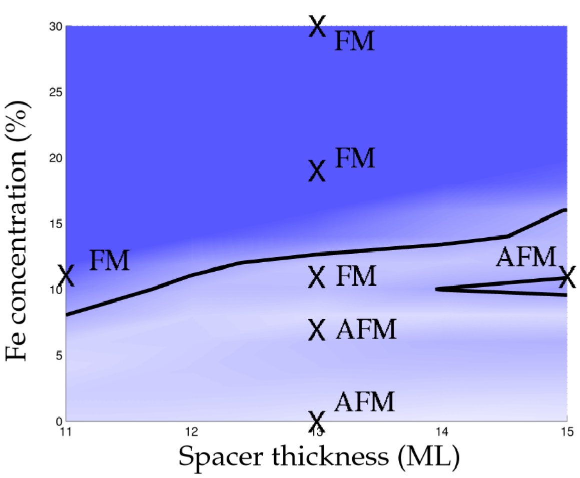

FIG. 3 (color online). Magnetic phase diagram for $Fe_3/[Fe_xV_{1-x}]_n$. The calculations include interface intermixing of $\Gamma_C = 1.8$ and a roughness of $\Gamma_T = 1.35$ in close analogy to Ref. [31]. The strength of the calculated IEC is represented by the blue scale (gray scale in the black and white version of the figure) where dark shades represent FM coupling and light shade represents AFM coupling. The black lines represent the border between the theoretical FM and AFM solutions. The experimental points are marked by X. The theoretical phase diagram was obtained by a linear interpolation of calculations for V thicknesses in steps of integer ML and Fe concentrations in steps of 2%. The amplitudes of the AFM samples with our choices of intermixing and roughness are $Fe_3V_{13}(0\%)$ 0.3115 mJ/m^2, $Fe_3V_{13}(7\%)$ 0.1773 mJ/m^2, and $Fe_3V_{15}(11\%)$ 0.0055 mJ/m^2.

ideal interfaces [26,27]. The extent of interface roughness and interface intermixing in the experimental samples is not known exactly. The chosen values are those found from previous studies of Fe/V interfaces in Ref. [25] and must be seen as an estimation. Our calculated amplitudes agree with experiments to a much better degree than if the interface alloying and roughness were not modeled (cf. captions of Figs. 1 and 3.) Any accumulation of either Fe or V at the interfaces has not been taken into account. Figure 3 shows that for low concentrations of Fe in the V spacer layer, the calculated magnetic phase diagram is complex, being dependent on both spacer layer thickness and Fe concentration. In particular for the chosen values of intermixing and roughness we conclude that at a spacer layer thickness of 14–15 ML the IEC should display a trend AFM → FM → AFM → FM, with increasing Fe concentration in the spacer layer.

The most interesting region of Fig. 3 is where the AFM coupling switches over to FM coupling. The mechanism is as described above due to direct exchange that dominates the AFM RKKY coupling. For spacer layer thicknesses around 13 ML, particularly favorable conditions are found due to that the dependence on spacer thickness of the switching is very weak (the borderline is almost flat). In addition, there are no interfering Fermi surface effects (as for 14–15 ML thicknesses) and the 13 ML V thickness system is very well characterized (it is known experimentally that the 0% Fe concentration point in Fig. 3 is AFM). Figure 3 also shows that theory and experiment agree on the sign of the interlayer interaction in all cases but one; the sample $Fe_3/[Fe_{0.11}V_{0.89}]_{13}$ has FM coupling in the measurements whereas theory puts this system in the AFM region. Inspection of Fig. 3 shows that a slight increase in Fe concentration would make the coupling FM also in the calculations.

Since the 13 ML region seems to be the most favorable we have chosen this region for our transport calculations. The results for $Fe_3/[Fe_xV_{1-x}]_{13}$ are listed in Table I, and it may be seen that the CPP-GMR effect is large for Fe/V multilayers and that the CPP-GMR is reduced with increasing Fe alloying of the spacer layer. The current-in-plane GMR was not calculated but can be expected to be a factor 2–3 lower than the CPP-GMR [28].

The calculated trends of the transport properties can easily be understood from the band structure of bcc Fe and bcc V. The minority spin bands of Fe and the bands of V match well [29], which results in good conduction for the minority spin channel. In contrast, the majority spin channel of Fe has poor matching with the V bands, which should result in poor conductance for the majority spin channel of the FM configuration and poor conductance for both channels of the AFM configuration. These expectations are shown in Table I to hold.

Alloying reduces the matching of the minority spins of Fe with V, which reduces the conductance. On the other hand, the conductance of the AFM configuration's channels is increased. This increase originates in the appearance

TABLE I. Calculated conductance (in e^2/h) for FM and AFM configurations of $Fe_3/[Fe_xV_{1-x}]_{13}$ and the corresponding GMR values (pessimistic ratio) given in percent. In parentheses we show the specular, $k_{\|}$-conserving, contribution to the total conductance (in percent). The diffusive, (non-$k_{\|}$-conserving) contribution is therefore given by the difference between the total conductance and the specular contribution.

x	FM ↑	FM ↓	AFM ↑	AFM ↓	GMR
0.00	0.147(100)	0.462(100)	0.138(100)	0.138(100)	54.7
0.04	0.134(46)	0.416(74)	0.167(43)	0.166(43)	39.5
0.08	0.128(37)	0.387(67)	0.166(33)	0.164(33)	35.9
0.12	0.122(34)	0.366(63)	0.166(29)	0.165(30)	32.2
0.16	0.121(29)	0.350(61)	0.166(25)	0.167(26)	29.3

of a diffusive, non-$k_{\|}$-conserving contribution with disorder that over compensates the decrease of the (low) specular $k_{\|}$-conserving contribution [17,30]. The combined effect of the decreased FM conductance and the increased AFM conductance leads to a reduced difference in conductance for the two configurations and consequently a decrease of the resulting GMR ratio (see Table I).

In summary, we have investigated the additional degree of freedom given by the introduction of a competing ferromagnetic direct exchange mechanism in antiferromagnetically coupled multilayers by alloying the spacer material with a magnetic impurity. The resulting necessary existence of a transition from antiferromagnetic coupling to ferromagnetic coupling means that we can tune the interlayer exchange coupling to a very low value. Our theoretical transport calculations indicate that it is possible to maintain a high GMR value when the transition is approached. It is quite likely that the alloying concentration is a better parameter to use for tuning the interlayer exchange coupling, since it is much easier to control than for instance the thickness of multilayers and spin-valves, where, in particular, the structural properties (roughness and mixing) of the interfaces are difficult to control.

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