# Oscillatory Exchange Coupling of Ferromagnetically Aligned Fe(110) Layers through Ag(111) Interlayers

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The interlayer exchange coupling in Fe(110)/Ag(111) multilayer structures with parallel alignment of the Fe layer magnetizations has been investigated with a zero-field Mössbauer spectroscopy technique that is sensitive only to the spin-wave spectrum at the Fe/Ag interface. Model spin-wave calculations are presented to connect the coupling strength to the observed interfacial spin-wave behavior. As the Ag thickness was varied from 0 to 40 ML (monolayers), the interfacial spectra of the films showed oscillations in the coupling strength with a period of 6 ML, in close agreement with recent predictions.

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Since the discovery of antiferromagnetic interlayer exchange coupling in Fe/Cr/Fe sandwich structures [1] and the subsequent discovery of oscillations in antiferromagnetic (af) coupling strength in Fe/Cr multilayers and many other similar structures, many workers have devoted considerable attention to these phenomena, experimentally [2–6] as well as theoretically [7–11]. More recently, the nature of the coupling has been clearly shown to be dependent on the structure, orientation, and Fermi-surface topology of the interlayer material [12]. In most cases, the oscillatory component of the interlayer coupling is strong enough to cause antiferromagnetic alignment of spins in adjacent magnetic bilayers, making detection of the af coupling possible by magnetometry. However, if the interlayer coupling strength is comparable to the anisotropy energies of the system, it is possible to have weak af oscillations or ferromagnetically (fm) coupled layers in which the fm coupling strength oscillates. In particular, the (111)-oriented noble metal spacers appear to exhibit very weak af coupling due to the high Fermi surface curvature near the external Fermi wave vectors. These weak oscillations may go undetected by a magnetometry technique because of the requirement of an applied field. In the case of Cu(111), observations of more than one af oscillation have proved difficult [13], while no af coupling has yet been reported in Ag(111). In this Letter we report the first evidence of oscillations in coupling strength through Ag(111) interlayers.

One method of directly obtaining information about the interlayer exchange coupling in ferromagnetically coupled multilayers is to measure the spin-wave spectrum. This can be done by various light scattering techniques [14], or by $^{57}$Fe Mössbauer spectroscopy in Fe-containing samples. For a three-dimensional ferromagnetic sample in the spin-wave region, the temperature dependence of the hyperfine magnetic field $H_{HF}$ at the Fe nucleus is expected to follow a Bloch $(1-BT^{3/2})$ law similar to the temperature dependence of the magnetization [15]. The quantity $B$, called the spin-wave stiffness parameter, is related to the exchange interaction experienced by the Fe spins. For thin film multilayers, the Bloch law will hold at low temperatures if the magnetic layer thickness is large enough, or if the interlayer exchange is strong enough that the coupled layers essentially form a three-dimensional system.

This group has previously demonstrated the existence of exchange coupling in the case of thin layers for Fe(110)/Ag(111) multilayers [16,17]. In the work in Refs. [16,17], we inferred the existence of interlayer exchange coupling from the observation of a dimensional crossover in the spin-wave spectrum as the interlayer thickness was varied from 4 to 20 atomic layers. As the interlayer thickness decreased, the $H_{HF}$ temperature dependence changed from quasilinear in $T$ (2D spin waves) to $T^{3/2}$ (3D spin waves). Because of this dimensional crossover, it was not possible to detect oscillations in interlayer coupling strength.

In the present work, a slightly different approach to the measurement of the spin-wave spectrum is required. To detect oscillations in the interlayer coupling, it is necessary to compare the spin-wave spectra of many different samples with the same dimensionality of magnetic behavior. We therefore must ensure that all the samples are three-dimensional, and obtain the spin-wave spectrum only in the region near the Fe/Ag interfaces. Fe atoms in this interfacial region will follow a Bloch law with a surface spin-wave stiffness parameter $B_S$ which is larger than that of the bulk. As the interlayer thickness in-

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creases from zero, and the interlayer exchange coupling $J_1$ falls off to zero asymptotically, $B_S$ approaches a value of 2 times $B$ [18]. If there are any oscillations superimposed on the asymptotic decrease in $J_1$, they will also be reflected in $B_S$. Calculations of $B$ from $J_1$ have previously been performed for bulk spin-wave properties of magnetic multilayers by Politi and co-workers [19] and Qiu *et al.* [20].

To model our system we begin with the Hamiltonian for two semi-infinite, simple cubic, Heisenberg ferromagnets, interacting through the interfacial ($l=-1,0$) planes via a nearest neighbor exchange $J_1$,

$$H=-\sum_{l,m\geq 0}\sum_{l_\parallel,\mathbf{m}_\parallel} J(l-\mathbf{m})\mathbf{S}_l\cdot\mathbf{S}_\mathbf{m} - \sum_{l,m\leq -1}\sum_{l_\parallel,\mathbf{m}_\parallel} J(l-\mathbf{m})\mathbf{S}_l\cdot\mathbf{S}_\mathbf{m} - 2J_1\sum_{l_\parallel}\mathbf{S}_{l_\parallel,0}\cdot\mathbf{S}_{l_\parallel,-1}\,, \tag{1}$$

where $l=(l_\parallel,l)$ are the lattice vectors and $J(l-\mathbf{m})=J_0>0$ for nearest neighbor pairs within each semi-infinite ferromagnet and zero otherwise. In the framework of spin-wave theory, we perform the usual linearized Holstein-Primakoff transformation from spin to bosonic operators and, owing to the lack of translational symmetry perpendicular to the interface, we work in the mixed Bloch-Wannier representation. In the case of ferromagnetic interfacial interaction ($J_1>0$), following Yaniv [21], we obtain in a direct way the interaction two particle Green's function, $G_{lm}(E,\mathbf{k}_\parallel)\equiv\langle\langle a_{\mathbf{k}_\parallel,l};a^\dagger_{\mathbf{k}_\parallel,m}\rangle\rangle_E$ from the noninteracting Green's function, by applying the interfacial part of the quadratic bosonic Hamiltonian as a perturbation on the noninteracting part, via Dyson's equation. In the case of antiferromagnetic interfacial interaction ($J_1<0$), we must introduce two different families of bosonic operators to take care of the opposite quantization directions for $l\leq -1$ and $l\geq 0$. We then obtain two Green's functions, $G_{lm}(E,\mathbf{k}_\parallel)$ and $G'_{lm}(E,\mathbf{k}_\parallel)\equiv\langle\langle a^\dagger_{-\mathbf{k}_\parallel,l};a^\dagger_{\mathbf{k}_\parallel,m}\rangle\rangle_E$, coupled via two Dyson's equations. In general, the spin-wave density of states at any plane $l$ is then expressed in terms of the trace over $\mathbf{k}_\parallel$ of the imaginary part of $G_{ll}(E,\mathbf{k}_\parallel)$,

$$\rho_l(E)=\frac{a^2}{\pi}\int\frac{d\mathbf{k}_\parallel}{(2\pi)^2}\,\mathrm{Im}G_{ll}(E,\mathbf{k}_\parallel)\,, \tag{2}$$

and the local magnetization at the plane $l$ is

$$\langle S^z_l\rangle=S-\int_{-\infty}^{+\infty}dE\,f(E,T)\rho_l(E)\,, \tag{3}$$

where $f(E,t)$ is the Bose-Einstein distribution. For ferromagnetic interfacial interaction, we find that the effective Bloch law,

$$\langle S^z_l\rangle/S=1-k(J_1/J_0)B_{sc}T^{3/2}\,, \tag{4}$$

is always followed, with the limits $k(1)=1$ and $k(0)=2$ for a single interfacial plane ($l=0,-1$). Here $B_{sc}$ is the value of $B$ in the Bloch $T^{3/2}$ law for a simple cubic ferromagnet. For antiferromagnetic interlayer exchange, in the limiting case $|J_1|=J_0$, the density of states at the interface turns out to be $\rho_0(E)\propto E$ and therefore the magnetization follows a $T^2$ law. Nevertheless, when $|J_1|$ decreases, the interfacial magnetization is found to follow an effective $T^{3/2}$ law even in the antiferromagnetic coupling case. In conclusion, in the temperature range probed by the experiments, the interfacial magnetization can be fitted by the effective Bloch law (4) provided that $-0.2\leq J_1/J_0\leq 1.0$. The prefactor $k(J_1/J_0)$ has been calculated for the valid range of $J_1$ using both the $l=0$ and the $l=1$ planes, and is shown in Fig. 1. It is worthwhile to note that in this figure $k(0)\neq 2$ because of the inclusion of the $l=1$ plane.

To look experimentally at the spin-wave spectrum at the Fe/Ag interfaces only, the 2 monolayers (ML) closest to the interface were composed of enriched (95.7%) $^{57}$Fe, while the rest of the Fe bilayers were natural Fe. In this arrangement, $\approx 92\%$ of the $^{57}$Fe in the sample is segregated to the interfacial region. Since the Mössbauer effect is sensitive only to the presence of $^{57}$Fe, the hyperfine parameters obtained correspond to the Fe within 2 ML of the Fe/Ag interface. Information about the interlayer coupling can therefore be obtained by measuring the $T^{3/2}$ prefactor ($B_S=kB$) in the Bloch law. Interdiffusion of the $^{57}$Fe and natural Fe would result in modified spin-wave characteristics in the case of zero coupling, as a result of the fact that we would then be measuring bulk properties in addition to the interface. Similarly, we would expect to see broadened Mössbauer lines. We conclude from the absence of these effects that such interdiffusion is not a significant effect under our growth conditions. Hysteresis properties for several of the samples were measured by vibrating sample magnetometry (VSM) at 4.2 K in order to determine the sign of the $J_1$ at the minima of $k(y)$. These total-moment measurements showed no unambiguous evidence of af coupling.

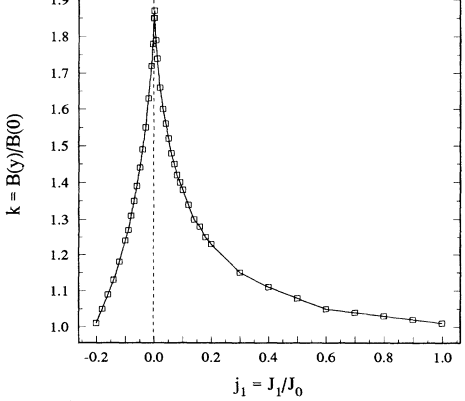


FIG. 1. The surface spin-wave stiffness parameter $k=B(y)/B(0)$ as a function of $j_1=J_1(y)/J_0$. The calculation is performed for the $l=0$ and $l=1$ layers of a simple-cubic biferromagnetic interface.

The hysteresis loop at $y=6$ ML has a stepped structure which is suggestive of very weak af coupling, but the Fe layers were clearly not aligned antiparallel, as the moment was nonzero at zero field. For samples not near $y=6$ ML the loops are completely fm in character. We conclude from the VSM measurements that although $J_1$ may be negative, it is too weak to overcome the anisotropies of the thick Fe(110) layers.

All the samples were prepared by molecular beam epitaxy (MBE) in a Perkin-Elmer PHI 430B MBE system equipped with *in situ* reflection high-energy electron diffraction (RHEED). The base vacuum in this system is $<2\times10^{-10}$ Torr, with the vacuum during growth $<2\times10^{-9}$ Torr. The multilayer structures all had the form $[\mathrm{Fe}(110)_x\,{}^{57}\mathrm{Fe}(110)_2\mathrm{Ag}(111)_y]_z+\mathrm{Fe}(110)_x$. In this study $x$ was always at least 20 ML, and the interlayer thickness $y$ was varied between 0 and 40 ML. The additional $\mathrm{Fe}(110)_x$ layer was grown to prevent the last $^{57}$Fe probe layer from existing as a free interface. Each multilayer structure was grown on a 5000 Å thick Ag(111) single crystal, which was grown *in situ* by MBE on a V-2 quality Mica substrate. Fe layer thicknesses were measured by quartz crystal microbalance, independently calibrated by profilometry, and Ag layer thicknesses were monitored by Electron Impact Emission Spectroscopy (Inficon Sentinel III). The flatness, crystallinity, and orientation of each Ag(111) substrate were investigated with RHEED before each multilayer growth. The RHEED patterns obtained from the substrates always showed sharp streaks with many orders visible, which is indicative of high quality, flat single crystals.

RHEED patterns obtained from the $^{57}$Fe(110) surfaces prior to the deposition of Ag suggest high quality, flat surfaces at the atomic scale. The Ag(111) pattern is very characteristic of diffraction from a two-dimensional surface lattice, and the Fe(110) pattern is only slightly rougher than that of the Ag(111) substrate. After the Ag interlayer is deposited on the $^{57}$Fe, the RHEED pattern improves almost to the same flatness as the Ag(111) substrate, indicating a highly repeatable structure. Considerable structural information can also be obtained from the transmission Mössbauer spectra, in addition to the magnetic information. A lack of line broadening in these surface-probed samples indicates that the surface being probed is very flat at the atomic scale. Any roughness at this length scale would result in a shallow composition gradient across the interface, and therefore a broad distribution of hyperfine magnetic fields at the $^{57}$Fe nuclei. In previous publications, we have reported on the question of Fe(110) growth on Ag(111), and typical RHEED patterns and Mössbauer spectra can be found in these reports [17,22].

The hyperfine fields of all the samples followed the Bloch $T^{3/2}$ law very closely. The values for $k(y)$ are plotted as a function of interlayer thickness $y$ in Fig. 2(a). The parameter increases to 1.83 with increasing interlayer thickness because of the overall weakening exchange, but has a clear oscillatory component superimposed on it. The minima of $k(y)$ occur at $y=6$, 11, and 18 ML, which is in close agreement with the single periodicity of 5.94 ML predicted by Bruno and Chappert for coupling through Ag(111) [10] based on the extended RKKY theory. From the spin-wave theory presented above, an asymptotic value of $k=1.86$ for zero coupling is expected. Such agreement between an idealized model [a sc (100) biferromagnetic interface with no roughness], and a real system [bcc Fe(110)/fcc Ag(111) multilayers] is unexpected, and suggests that the essential physics is in fact contained in the simple-cubic spin-wave model. In attempting to reconstruct the observed behavior of $k(y)$ from the theoretical predictions, we must start with an oscillating $J_1(y)$. Using the RKKY function obtained in Ref. [10],

$$J_1(y)=-J_0\frac{d^2}{y^2}\frac{m^*}{m}\sin\left(\frac{2\pi y}{\Lambda}+\phi\right)\frac{y/L(T)}{\sinh[y/L(T)]}, \quad (5)$$

values for $k(y)$ can be obtained by the spin-wave calculations described in this Letter. Here we use $J_0$ as the energy scale for $J_1(y)$, and $\Lambda$, $m^*$, and $L$ are treated as parameters. In this treatment we take $\phi=\pi/2$ as obtained

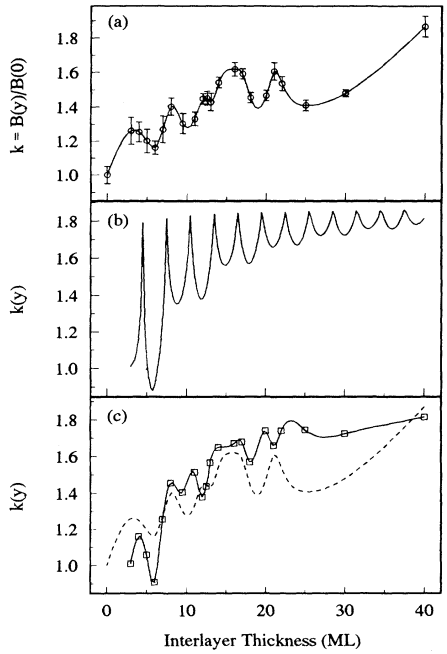


FIG. 2. Oscillatory interlayer coupling in $[\mathrm{Fe}(110)_x\text{-}{}^{57}\mathrm{Fe}(110)_2\mathrm{Ag}(111)_y]_{15}+\mathrm{Fe}(110)_x$ multilayer structures. (a) Measured surface spin-wave stiffness parameter for interlayer coupling through Ag(111). (b) $k(y)$ as predicted by fitting extended RKKY theory from Ref. [10] to the data. (c) Sampling of predicted $k(y)$ values at points where experimental data exist. In (c), the dashed line represents the experimental data, while the solid line with boxes is the sampled fit. For all samples $x\geq 20$ ML, sufficient to cause three-dimensional spin-wave behavior.

in Ref. [10] and note that the RKKY treatment does not predict correct phases in metallic multilayer systems. For Ag(111) interlayers, the fit parameters have predicted values of $\Lambda = 5.94$ ML, $m^*/m = 0.17$, and $L = 5.54$ ML at 300 K [10], however, the important quantity to test the RKKY theory is $\Lambda$, the oscillation period. In Fig. 2(b), we plot $k(y)$ obtained by performing a least-squares fit of $k(y)$ to our data. $k(y)$ is extremely sensitive to very weak interlayer couplings, due to the steepness of $k$ vs $J_1$ near $J_1 = 0$. The surface spin-wave stiffness parameter must have its maximum value of $1.86B$ at every point where $J_1(y)$ passes through zero. It is this feature of the spin-wave characteristics which makes this surface sensitive method highly suitable for the detection of coupling strength in very weakly antiferromagnetic multilayer structures, in addition to ferromagnetic structures. Since the peaks in $k(y)$ are very sharp, it is easy to miss them even when sampling at 1 ML intervals. Figure 2(c) illustrates what happens when the $k(y)$ predictions obtained from Eq. (5) are sampled at only the interlayer thicknesses where we have data. The solid line represents a least-squares fit of this sampled function to the data. The qualitative agreement to the data is quite good; at all the dips in our data up to $y = 22$ ML, a corresponding dip occurs in the best fit very close to the same interlayer thickness. The overall magnitude of the oscillations is predicted to be larger than observed, which is the cause of the apparent dip below $k = 1.0$ for $y = 6$ ML. In this region $J_1 < -0.2J_0$, which is not in the region of validity for Eq. (4), and a $T^{3/2}$ temperature dependence should not be seen. Since all the films even in this region do follow a Bloch law, we conclude that $J_1$ is within the valid region for Eq. (4). It is important to note that although $m^*$ and $L$ for the best fit are not close to theoretical predictions, $\Lambda$ is very close at 6 ML. Since we have fitted the Mössbauer data to an RKKY function which exhibits strong switching between af and fm coupling, agreement of the predicted amplitudes with observed values would be unexpected. We have shown, however, that the predicted period for Ag(111) is very close to the experimental value, and this is the critical test of the RKKY theory.

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