Reversible Tuning of the Magnetic Exchange Coupling in Fe/V (001) Superlattices Using Hydrogen

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The interlayer ordering between ferromagnetic Fe layers in Fe/V (001) superlattices is switched from initially parallel to antiparallel, as well as antiparallel to parallel, upon introducing hydrogen to the V layers. This process is reversible upon removal of the hydrogen. The results unambiguously prove that the major cause of the interlayer coupling transitions is not the hydrogen-induced changes of the thickness of the V layers, but most likely the distortion of the Fermi surface in the V layers. [S0031-9007(97)03652-1]

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Since the discovery of the oscillatory magnetic ordering in metallic multilayers [1], substantial work has been devoted to the exploration of the underlying mechanism of the interlayer exchange coupling. For transition metal superlattice systems, current interpretations attribute the basis of the coupling to extremal values of the Fermi wave vector and the discrete lattice spacing of the constituents [2–4]. In these models, the coupling between ferromagnetic layers oscillates between parallel and antiparallel alignments as a function of the thickness of an intervening spacer layer. Although a conceptually attractive description of the nature of the exchange coupling is obtained, the complexity of the Fermi surface makes the interpretation far from trivial for the transition metals. The detailed topography of the Fermi surface is expected to play a vital role in the exchange coupling [5]. Experimental exploration of this fundamentally important issue is difficult, since the means to continuously alter the lattice spacing and/or the Fermi energy are limited. One of the first attempts to systematically investigate these effects was on (110) $\mathrm{Fe}/\mathrm{Cr}_{1-x}\mathrm{V}_x$ multilayers [6]. The experimental results were successfully reproduced theoretically, but no experimental data on antiferromagnetic (AFM) coupled Fe/V (001) multilayers were presented.

AFM ordered Fe/V (001) superlattices have only recently been produced and characterized [7,8]. Samples with three monolayers of Fe were found to be magnetically isotropic in the plane, independent of the V layer thickness [8]. If the electronic states and/or the thickness of the V layers of these samples could be altered continuously, an attractive way of exploring the details of the magnetic interlayer coupling between the ferromagnetic Fe slabs across the nonmagnetic V would be obtained. We will show that this can be done by loading the V layers with H.

The hydrogen uptake of Fe/V superlattices is well known, and the H is found to reside exclusively in the V lattice [9]. The total thickness of the V layers can be changed reversibly by as much as 10% at moderate H pressures, without any memory effects [10]. Ignoring the changes in the electronic structure, one can view the introduction of hydrogen as a way to reversibly change the thickness of the V spacer layer, and thereby alter the coupling between the Fe layers. In Fig. 1, we have plotted the measured saturation field of Fe/V (001) superlattices for different thicknesses of the hydrogen free V layers. The tunable V thickness range which is accessible in each sample by the hydrogen loading, is indicated by arrows in the figure. Solely considering the effects of the expansion of the V layers on the coupling between the Fe layers, the introduction of H would enhance the AFM exchange coupling in the Fe(3)/V(12) sample, whereas in the Fe(3)/V(14) and Fe(3)/V(15) samples it would diminish. [We will use the shortened notation, Fe(3)/V(*n*), where the number in the parentheses refers to the closest number of monolayers. Fe(3)/V(12) denotes therefore three monolayers of Fe separated by twelve monolayers of V.] However, if there are changes in the electronic structure, the above description would be incomplete, which we will indeed show below.

The Fe/V (001) superlattices were grown on $20 \times 20 \times 1\ \mathrm{mm}^3$ epi-MgO substrates, as described in

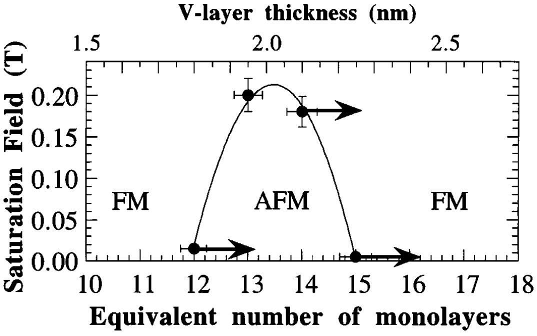

FIG. 1. Saturation field as a function of the V layer thickness for AFM coupled Fe(3)/V(*x*) superlattices, obtained by the magneto-optic Kerr effect and SQUID measurements. The thicknesses which are accessible by loading the samples with hydrogen are marked by arrows in the figure.

Ref. [7]. Typical numbers of the double layers and total thickness were 70–80 and 180–200 nm, respectively. The samples were covered with 5 nm Pd to prevent oxidation and to ensure fast hydrogen uptake and release. Reflectivity and high-angle x-ray diffraction analysis revealed good crystal quality, well-defined repeat length, and sharp interfaces.

The measurements were performed on the NG1 reflectometer at National Institute of Standards and Technology, in the molecular beam epitaxy chamber for *in situ* neutron scattering which also has H-loading capabilities. Measurements were made at room temperature in a field of ≈100 G, under hydrogen pressures from 0.1 to 10^5 Pa and in ultrahigh vacuum. The samples were initially heated to ≈400 K to remove passivating adsorbates from the surface. The kinetics were sufficiently fast to allow one to load and unload the samples at room temperature. However, at low pressures, the kinetics were slow enough to allow several measurements at different hydrogen concentrations as the sample approached equilibrium.

The magnetic interlayer ordering can be directly monitored using spin polarized neutron reflectivity [11]. In elastic specular neutron reflectivity measurements, the neutron wave vector transfer $\mathbf{Q} = \mathbf{k}_f - \mathbf{k}_i$ is parallel to the film's surface normal ($\mathbf{k}_f$ and $\mathbf{k}_i$ are the reflected and incident wave vectors, respectively). Analyzing these data can yield the scattering-length density profile along the surface normal direction of the film, which can in turn be interpreted as the chemical composition profile. Furthermore, for polarized neutrons the reflectivity is spin dependent and allows one to determine the magnetization profile in the sample as well. As depicted in the inset of Fig. 2, incident neutrons can be polarized in one of two states, $+$ or $-$, and can be scattered with or without change of state. Thus four cross sections can be measured, $(+,+)$, $(+,-)$, $(-,+)$, and $(-,-)$, where the two signs refer to the incident and scattered polarization states. The non-spin-flip (NSF) scattering is due to a combination of the chemical scalar scattering potential and the component of the magnetic vector scattering potential perpendicular to $\mathbf{Q}$ and along the polarization axis. The $(+,+)$ scattering depends upon the sum of these potentials, whereas the $(-,-)$ scattering depends on their difference. The spin-flip scattering (SF) arises from the component of the in-plane magnetization perpendicular to the polarization axis. For a superlattice structure, peaks in the scattered intensity will be found at $Q_{\mathrm{FM}} = 2\pi/\Lambda$ corresponding to the superlattice repeat distance Λ. At half this Q_{FM}, a peak in the scattered intensity would correspond to a structure with twice the period of the chemical modulation, e.g., an AFM structure. Therefore one can deduce the total magnetization in the Fe layers from the splitting between the $(+,+)$ and $(-,-)$ cross sections at the ferromagnetic (FM) peak position, and the degree of AFM order from the SF scattering into the half-order peaks. After correcting for instrumental effects such as the efficiencies of the neutron polarizers, and the presence of a small fraction of neutrons with wavelengths of a higher harmonic ($I_{\Lambda/2}/I_{\Lambda} = 0.0478$), we found that only spin-flip scattering remains at the half-ordered peaks, indicating that in the AFM ordering, the Fe layers are oriented perpendicular to the applied field.

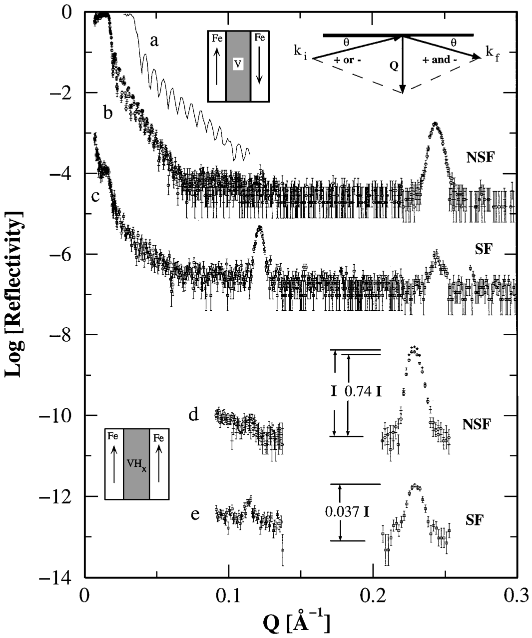

FIG. 2. Neutron scattering data for Fe(3)/V(14) virgin and hydrogen-loaded samples. Observe that the data are presented on a log scale. These results were obtained at 100 G and room temperature. The inset in the upper right corner shows the neutron reflection geometry for the polarized beam. Data set "*b*" is the $(+,+)$ and $(-,-)$ components whereas "*c*" is the combined $(+,-)$ and $(-,+)$ components, from the hydrogen-free sample. The curve labeled "*a*" is the $(+,+)$ intensity of curve "*b*," expanded by a factor of 2 along the *x* axis to show the finite-size oscillations. "*d*" and "*e*" correspond to "*b*" and "*c*," for the same sample exposed to 14 kPa hydrogen pressure. The other insets depict the magnetic alignment of the Fe layers for the corresponding data. Data sets "*c*," "*d*," and "*e*" are displaced downward by 2, 6, and 8 orders of magnitude, respectively, for clarity.

A typical reflectivity curve for the Fe(3)/V(14) sample, with and without H, is shown in Fig. 2. For clarity we display only the $(+,+)$, $(-,-)$, and the combined spin-flip components, since $(+,-)$ and $(-,+)$ are identical. As seen in Fig. 2, a number of rapid oscillations are present at low Q values, which originate from the overall thickness of the sample. When loaded with H the half-order peak gradually diminishes and eventually vanishes, while the NSF component of the FM peak splits due to the net magnetization of the ferromagnetic alignment of the Fe layers. In addition, while the SF component of the FM peak appears to increase upon H loading, it remains approximately the same fraction of the NSF component in both the H-loaded and as-grown cases, and is not significantly different than what would be expected given the imperfect instrumental polarizing and flipping efficiencies.

In Fig. 3, we plot both the integrated intensity of the spin-flip components of the AFM peak and the difference between the NSF integrated intensity of the FM peaks for the Fe(3)/V(12), Fe(3)/V(14), and Fe(3)/V(15) samples, as functions of the relative shift of the first-order structural peak (Q_{FM}). The V lattice expands with increasing H content, which results in a decreasing Q_{FM}; hence the change in Q_{FM} directly indicates the amount of H in the sample. In these curves, the data were taken in no particular order, and all the repeated points gave the same integrated intensities regardless of the hydrogen-loading history, indicating complete reversibility without any hysteresis effects. An estimation of the H concentration is obtained from the relation between the H concentration and the lattice expansion [10]. In the Fe(3)/V(12) sample, the intensity of the AF component diminishes rapidly and eventually disappears at a shift of 0.5%, while the net magnetization increases. For the sample with 14 monolayers of V, higher H concentration is required to switch from AFM to FM ordering, corresponding to a 2% expansion of the V layers. In the Fe(3)/V(15) sample, no AFM peak is observed without H present in the sample. By introducing H in the V layers, an antiferromagnetic alignment of the Fe layers is obtained, which diminishes while increasing the H content further. The FWHM of this H-induced AFM resonance is only ≈ 0.01 in $|\Delta Q/Q|$, corresponding to an $\approx 5\%$ change in the hydrogen content. No hysteresis effects were observed.

The effect of H on the magnetic ordering in (110) textured Fe/Nb multilayers was recently addressed by Klose *et al.* [12]. The interlayer ordering between Fe layers was found to be altered by the presence of hydrogen in the Nb (AFM to FM ordering only), but a large hysteresis effect was present. Hydrogen-induced switching capabilities are therefore not only restricted to V-based superlattices.

If the main contribution to the change in the coupling in the Fe/V superlattices originated from the increase of the V spacer thickness, the Fe(3)/V(12) sample should show an increasing AFM coupling with H loading. However, the opposite was observed. We must therefore conclude that the main contribution to the change of the type of coupling does not originate from the increase in the V thickness. Rather, to overcome the expansion, an even faster increase of the oscillatory exchange coupling period is required. This change in period is most likely due to the effect of the H on the Fermi surface of the V layers. This is clearly seen in Fig. 4, where we plot the intensity of the AFM peak as a function of the V layer thickness. The thickness of the V layers for the Fe(3)/V(14) and Fe(3)/V(15) are overlapping, but the magnetic interlayer ordering is different because they have different hydrogen contents.

The influence of the hydrogen uptake on the band structure in bulk V is reasonably well known. However, no systematic investigations of the influence of the hydrogen content on the topography of the Fermi surface do

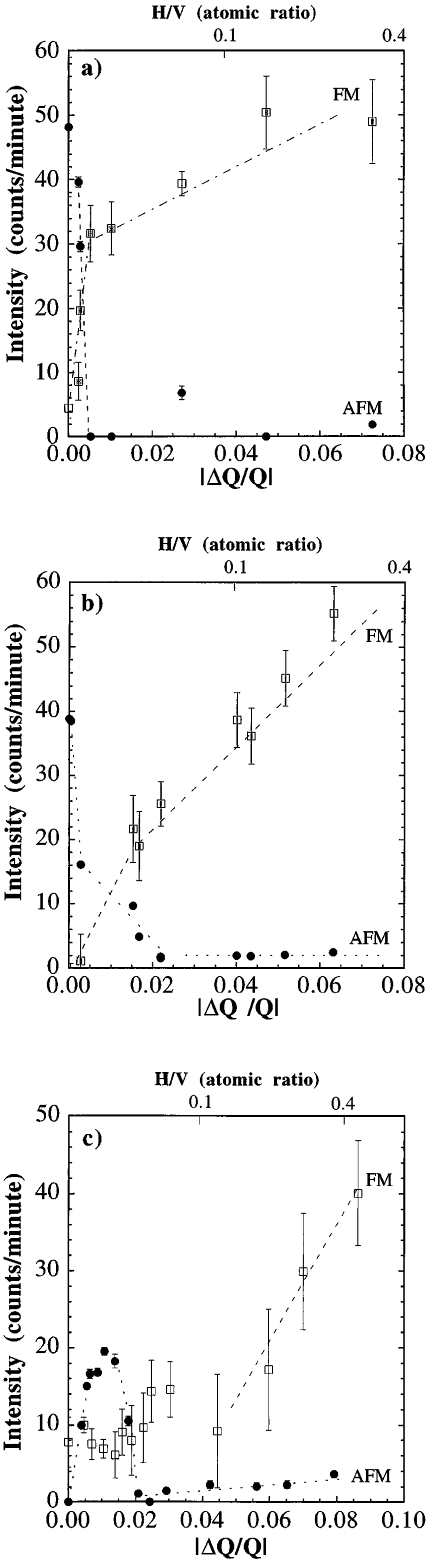

FIG. 3. The intensity of the spin-flip component from the AFM peak and the difference between the $(+,+)$ and $(-,-)$ scattering of the first-order peak at different hydrogen loadings for (a) Fe(3)/V(12), (b) Fe(3)/V(14), and (c) Fe(3)/V(15) superlattices. The hydrogen concentration is estimated from the measured lattice expansion, using the results from previous x-ray diffraction investigations. Dashed lines are to guide the eye.

exist. Furthermore, as the tetragonal distortion of the V in the superlattice structure is *gigantic*, a direct comparison with a bulk hydride would not be legitimate. The hydrogen-induced changes in the electronic structure of Fe-V superlattices have recently been studied theoretically by Coehoorn [13]. For ordered body-centered-tetragonal VH he found that the Fermi level is raised with respect to the 3D band to a position slightly above the deep valley in the density of states separating bonding and antibonding states. In this sense the hydrided V layers are therefore expected to become Cr-like. However, Schilfgaarde and co-workers concluded that the alloying of the V with Cr resulted in a decrease of both the short- and the long-period oscillations [6], which is the opposite of what we observed. Hence, the changes of the Fermi level alone do not give the essence of the observed change in the interlayer magnetic ordering; the detailed topography of the Fermi surface has to be considered.

We can estimate the change in the extremal values (ignoring the possible change in the scattering phase shift) using

$$J(d) = \sum_{\alpha} \frac{J^{\alpha}}{d^2} \sin(q_{\perp}^{\alpha} d + \phi^{\alpha}),$$

where J^{α}, d, $q_{\perp}^{\alpha}$, ϕ^{α}, and α are the exchange coupling, the thickness of the V layers, the critical spanning vector, the phase shift, and the sum over all critical points of the Fermi surface, respectively [5]. When introducing hydrogen in the Fe(3)/V(15) sample, the total switching from FM to AFM to FM occurs within an expansion of $|\Delta Q/Q| = 0.02$, which corresponds to only roughly $\frac{1}{3}$ of a monolayer. If the observed switching were due to a change in the V layer thickness alone, a contraction of 3 monolayers would have been required (see Fig. 1). Hence, the hydrogen-induced changes on $q_{\perp}^{\alpha}$ (or ϕ^{α}) must be roughly 10 times faster than the change which is observed by *only* changing the V thickness. This simple interpretation is also consistent with the AFM to FM transitions as more hydrogen is required to switch the Fe(3)/V(14) than the Fe(3)/V(12) sample.

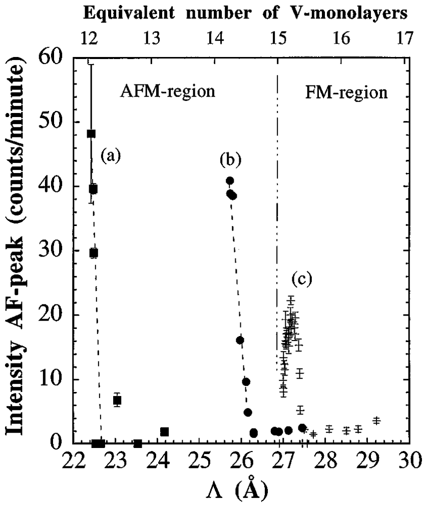

FIG. 4. The intensity of the spin-flip component from the AFM peak for all the samples under study, displayed as a function of the superlattice modulation period and the V layer thickness: (*a*) Fe(3)/V(12), (*b*) Fe(3)/V(14), and (*c*) Fe(3)/V(15). The measurements were performed at room temperature at a field of 100 G. Observe the overlapping in thickness of the hydrided and unhydrided samples. The regions labeled FM and AFM correspond to the magnetic ordering in H-free samples. Dashed lines are to guide the eye.

To summarize, the introduction of hydrogen is found to reverse the AFM interlayer ordering in Fe(3)/V(14) and Fe(3)/V(12) superlattices. The amount of hydrogen which is required to obtain a FM state in an external field of 100 G is only a few atomic percent. In the Fe(3)/V(15) superlattice, on the other hand, the introduction of H induces an AFM ordering in an extremely narrow H concentration range. In the latter two examples, the transition cannot be caused by the change in V thickness, since this requires contraction of the V layers. We therefore conclude that it originates from the modification of the Fermi surface in the V layers.

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