Exchange coupling through spin-density waves in Cr(001) structures: Fe-whisker/Cr/Fe(001) studies

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Exchange coupling through a spin-density wave in Fe-whisker/Cr/Fe(001) structures has been studied using Brillouin light scattering (BLS) and magneto-optical Kerr effect (MOKE). The Fe-whisker(001) substrates provide nearly ideal templates: they are characterized by atomic terraces having dimensions in excess of several micrometers. Such templates are essential for the study of short-wavelength exchange coupling which is mediated by the intrinsic spin-density wave in Cr(001). Atomically smooth Cr(001) layers similar to those of the Fe-whisker surfaces can be grown at raised substrate temperatures. Angular resolved auger electron spectroscopy measurements have shown that the Fe-whisker/Cr(001) interfaces are affected by an atom exchange placement mechanism (interface alloying). It will be shown that this interface alloying at the Fe-whisker/Cr interface profoundly affects the behavior of the short-wavelength oscillations. The phase of the short-wavelength oscillations is reversed compared to that expected for the spin-density wave in Cr(001). The strength of coupling is significantly decreased from that obtained from first-principles calculations, and the first crossover to antiferromagnetic coupling occurs at 4 ML. BLS and MOKE have shown unambiguously that the exchange coupling in Fe-whisker/Cr/Fe(001) structures can be described by bilinear and biquadratic terms. Experiments carried out using Cu and Ag atomic layers between the Cr(001) and Fe(001) films, i.e., heterogeneous interfaces, have shown that the exchange coupling in Cr(001) is strongly affected by electron multiple scattering. It will be argued that the exchange coupling through thick (>8 ML) and atomically smooth Cr(001) spacers can be described by localized interactions (Heisenberg type) and by electron multiple-scattering (quantum well state) contributions. This is in good accord with recent first-principle calculations by Mirbt and Johansson. However, interface alloying severely affects the behavior of the exchange coupling for Cr thicknesses less than 8 ML. In this thickness regime the overall coupling exhibits mostly a long-wavelength behavior with a small superimposed short-wavelength contribution. This initial Cr thickness regime is responsible for changes in the phase of the short-wavelength oscillations and for the reduced strength of the exchange coupling due both to the localized and to the multiple-scattering contributions. We have observed no significant dependence of the exchange-coupling strength on the Fe film thickness for samples having the structure Fe-whisker/11Cr/*n*Fe/20Au where *n* specifies an iron film thickness between 5 and 40 ML. However, preliminary data show that the exchange coupling is significantly increased in specimens for which both sides of the iron film are covered by Cr, i.e., for structures of the form Fe-whisker/11Cr/*n*Fe/11Cr/20Au. It appears that electron resonant states in the iron film play no important role in the strength of the exchange coupling when the iron is bounded on one side by the gold, but that they do become important when the iron film is bounded by Cr on both sides. BLS and MOKE studies on Fe-whisker/Cr/Mn/Fe(001) samples revealed that the antiferromagnetic state of Mn is composed of compensated (001) atomic planes. The results of the above experimental studies will be compared to recent theories. Points of agreement and of disagreement between the experimental results and recent first-principles calculations will be explicitly pointed out. [S0163-1829(99)10921-4]

INTRODUCTION

Fe-whisker/Cr/Fe(001) systems have played a crucial role in the study of exchange coupling between two ferromagnets separated by a nonferromagnetic spacer. Studies carried out by Unguris, Celotta, and Pierce[1] using scanning electron microscopy with polarization analysis (SEMPA), and the magneto-optic Kerr effect (MOKE) measurements by Purcell *et al.*[2] using Fe-whisker/Cr/Fe(001) samples showed that the exchange coupling oscillates with a short-wavelength period of ~2 monolayers (ML). The SEMPA images revealed in a very explicit way that short-wavelength and long-wavelength oscillations existed in the thickness range 5–80 ML of Cr. The period of the short-wavelength oscillations, $\lambda = 2.11$ ML, was found to be slightly incommensurate with the Cr lattice spacing; the period of the long-wavelength oscillations was found to be 12 ML. Short- and long-wavelength oscillations have also been observed by Grünberg and co-workers in Fe/Cr/Fe samples grown on a GaAs(001) substrate covered with a thick buffer layer of Ag(001).[3] Heinrich and co-workers (the SFU group) have carried out quantitative studies using Fe-whisker/Cr/Fe(001) samples.[4,5] The objective of the SFU group was to grow samples having the best available interfaces, to measure quantitatively the strength of the exchange coupling, and to compare these coupling strengths with *ab initio* calculations that explicitly include the presence of spin-density waves in the Cr. The requirement of smooth interfaces limited our study to samples which were grown on Fe-whisker templates with the Cr spacers terminated at an integral number of Cr atomic layers. It was found that the strength of the exchange coupling through the Cr(001) spacer is extremely sensitive to small variations in growth conditions. The measured exchange coupling was found to be reproducible only in those samples that exhibited layer by layer growth.

In our studies we concentrated on samples for which the Cr thickness ranged from 4 to 13 atomic layers. We wished to study a thickness regime ranging from a thin Cr interlayer for which the exchange coupling was dominated by the predominantly antiferromagnetic long-wavelength 12 ML dependence on thickness to a thickness regime in which the exchange coupling was dominated by the 2 ML short-wavelength period. In particular, we wished to investigate the origin of the deviation of the thickness dependence of the coupling from the predictions of simple Cr spin-wave theory (antiferromagnetic coupling for an even number of Cr atomic layers and ferromagnetic coupling for an odd number of Cr atomic layers). As will be discussed below, deviations from simple spin-wave theory can be ascribed to interface mixing of the Fe and Cr atoms.

GROWTH

Specimens were grown in an ultrahigh vacuum system (UHV) having a base pressure less than 10^{-10} Torr. Cr and Fe films were deposited on an iron-whisker(001) surface by evaporation from standard source ovens. The films were deposited at a rate of approximately 1 ML per minute at a pressure of $\sim 5\times10^{-10}$ Torr. Thicknesses were monitored using a standard quartz crystal gauge. The intensity of the reflection high-energy electron diffraction (RHEED) specular spot was measured during the film growth. We used a 10 keV electron beam incident on the specimen at a grazing angle (1 to 3 degrees). The specular spot intensity exhibited the periodic variations characteristic of layer by layer growth. The existence of unattenuated RHEED intensity oscillations of the specular spot during the growth of the Cr spacer did not guarantee reproducible values of the exchange coupling between a thin Fe(001) film and the whisker(001) substrate. It was necessary to establish conditions such that new layers were initiated at a repeatable pattern of nucleation sites. This condition could best be monitored by examining RHEED intensity oscillation amplitudes together with the width of the RHEED specular spot profile. The best results were obtained for the case when the RHEED intensity oscillations exhibited a cuspy, unattenuated pattern and the spot profile oscillated repeatably between narrow peaks (filled atomic layers) and wider, split intensity peaks (half filled layers); see Figs. 1(a) and (b). These requirements could be achieved by maintaining the substrate temperature in a narrow range of temperatures, $280\,°\mathrm{C} < T_{s,\mathrm{opt}} < 320\,°\mathrm{C}$. The presence of the specular spot splitting, see Fig. 1(b), in a direction parallel with the RHEED streaks for half filled atomic layers indicated that new atomic layers were formed from nucleation centers which were separated by a well defined mean distance. The observed splitting corresponded to a mean separation between atomic islands of ~800–900 Å at $T_{s,\mathrm{opt}}$.[4] This interpretation is in agreement with the scanning tunnel microscope (STM) studies by Stroscio *et al.*[6] Samples grown at the optimum temperature showed unattenuated RHEED intensity oscillations with well defined cusps at the RHEED intensity maxima; see Fig. 2. The first monolayer of Cr exhibited a unique behavior; the first RHEED intensity oscillation showed a strong peak with a very sharp cusp, see Fig. 2(a), even at substrate temperatures as low as 150 °C, indicating that the first atomic layer grows very smoothly. The situation changes when Cr is deposited on a Cr template. In that case the growth of Cr proceeds layer by layer only if the substrate temperature is adjusted to an optimum growth temperature $T_{s,\mathrm{opt}}$; see Figs. 2(a) and (b). In order to get a final Cr atomic layer nearly as smooth as the Fe-whisker template the substrate temperature can be changed gradually, but the approach to $T_{s,\mathrm{opt}}$ has to be from temperatures less than $T_{s,\mathrm{opt}}$. In a proper layer by layer growth the RHEED

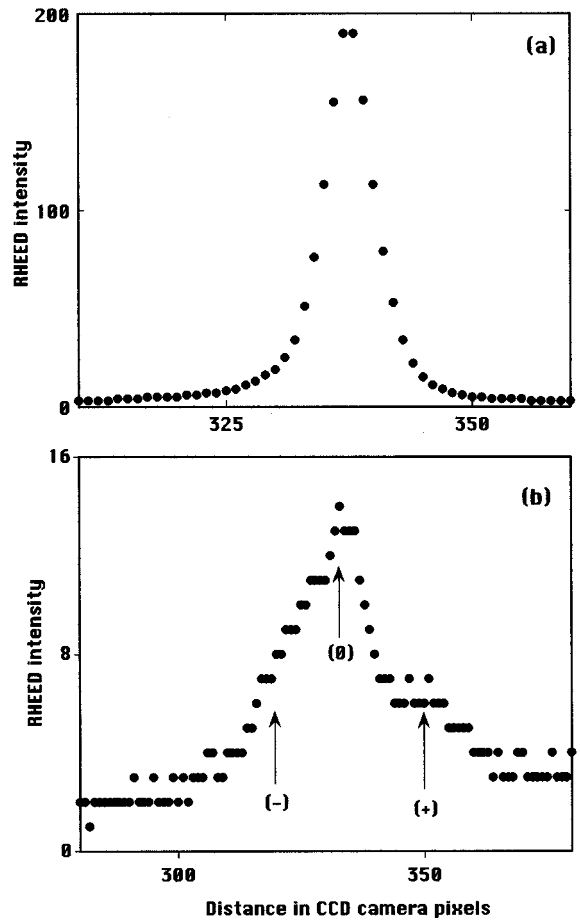

FIG. 1. Typical RHEED specular spot profiles for the growth of Cr on an Fe-whisker(001) template. The spot profile scan was taken along the direction which is determined by the intersection of the electron-beam plane and the plane of the fluorescent screen (in the direction of the RHEED streaks). The spot profiles were monitored at the second anti-Brag reflection corresponding to $2\theta\sim3$ degrees. (a) The spot profile at a maximum of the RHEED intensity oscillations, corresponding to a completely filled top atomic layer. The width of the specular spot profile is limited by the RHEED instrumental resolution. (b) The spot profile at a minimum of the RHEED intensity oscillations, corresponding to a half filled top atomic layer. The spot profile consists of the sum of three peaks: an attenuated version of the profile of (a) due to incomplete cancellation of the interference between electrons scattered from the different exposed surface levels (0), and two flanking peaks due to the presence of atomic islands (+) and (−). The splitting of the two flanking peaks determines the average lateral spacing of the Cr atomic islands. The average atomic spacing at $T_{s,\mathrm{opt}}$ is approximately 80–90 nm.

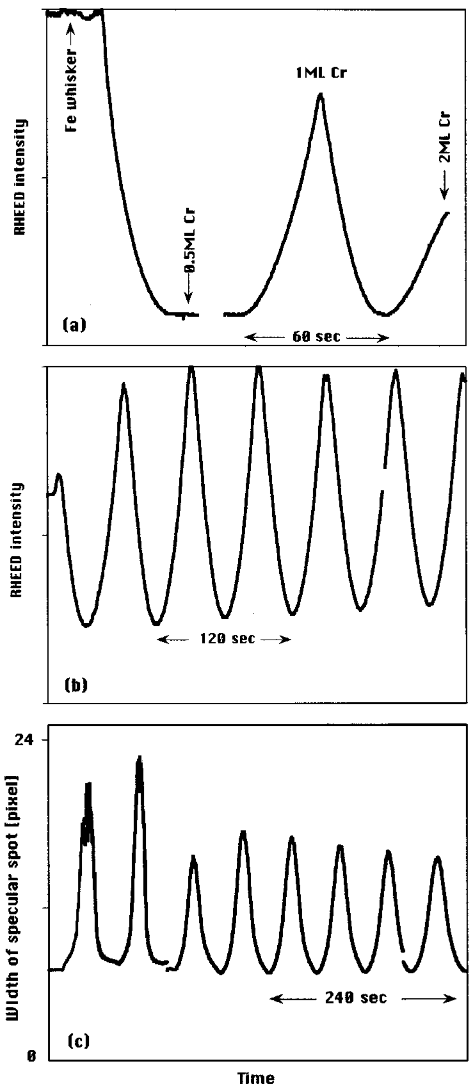

FIG. 2. The RHEED intensity oscillations (second anti-Brag reflection) of Cr on an Fe whisker(001) template. One period of oscillations, usually 60 seconds, corresponds to one additional atomic layer of Cr. (a) The first two oscillations at $T_s \sim 150\,°\mathrm{C}$. The measurement was halted at the peak of the second oscillation. Note that the second maximum is significantly lower compared to that for the top surface of the Fe whisker template and to that for the first filled atomic layer of Cr. (b) The substrate temperature was increased to establish layer by growth. The gradual increase of the RHEED intensity minimum is due to a gradual increase of the distance between the Cr nucleation centers during the growth: this was caused by an upward drift of the Fe whisker temperature. The size of the atomic islands was nearly the same as the lateral instrumental resolution of the RHEED gun and consequently one does not obtain a complete cancellation of the RHEED intensities at the half covered surfaces. The strength of the exchange coupling was found to be most reproducible for those samples where the minimum values of the RHEED intensity oscillations remained constant and were close to zero intensity. The growth in this figure was carried out at a somewhat higher temperature than $T_{s,\mathrm{opt}}$; see the text. (c) The full width at half maximum of the specular spot as a function of the Cr layer thickness for the RHEED intensity oscillations shown in (a) and (b). The maxima and the minima correspond to half and fully filled atomic layers, respectively. Note that the spot profile minimum widths are equal to that of the Fe-whisker template.

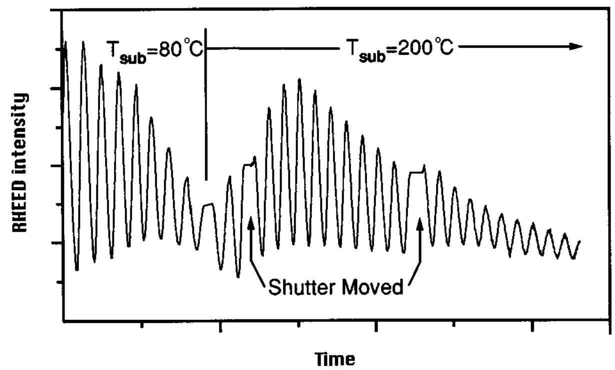

FIG. 3. The RHEED intensity oscillations (second anti-Brag reflection) for Fe growth on a Cr template. This growth was carried out for an Fe-whisker/Cr/Fe(001) sample having three different thicknesses for the deposited Fe layer. The growth was stopped, the shutter was moved, and the growth was again resumed as indicated in the figure.

intensity oscillations maintain a cuspy unattenuated pattern, see Fig. 2(b), and the width of the specular spot oscillates between minima (fully filled atomic layer) and maxima (half field atomic layer); see Fig. 2(c). The width of the specular spot corresponding to a complete atomic layer of Cr is as narrow as that of the Fe-whisker template. In order to obtain a Cr spacer having a well defined thickness one has to avoid excessive growth at naturally occurring atomic steps. The growth from such atomic steps results in the atomic steps moving across the surface. Thus the atomic steps on the Cr surface are displaced from those on the Fe-whisker substrate surface and the thickness of the Cr layers would necessarily fluctuate by 1 ML across the sample surface. This mechanism can partly provide an explanation for why samples grown outside the optimum range of substrate temperatures showed a large biquadratic exchange coupling and a small bilinear coupling.

The growth of Fe on a Cr(001) template proceeds very

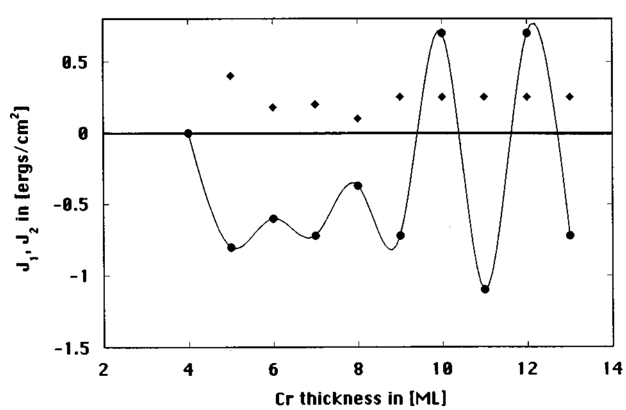

FIG. 4. The thickness dependence of the bilinear J_1 (●) and biquadratic J_2 (♦) exchange coupling. The biquadratic coupling can be measured only for AF coupled samples. The values of J_2 for the FM coupled samples (10, 12 ML) were assumed to be the same as that for the AF coupled samples containing 9, 11, and 13 ML of Cr. Note that the coupling becomes AF for thicknesses greater than 4 ML and the thickness dependence of J_1 has a broad AF maximum around 7 ML of Cr. Large short-wavelength oscillations appear for Cr thicknesses greater than 9 ML.

well for approximately 5–8 ML even at room temperature (RT). At the beginning of the growth the RHEED oscillations have a large amplitude even for the second anti-Bragg condition; see Fig. 3. The amplitude of the RHEED oscillations gradually decreases with increasing Fe layer thickness. The RHEED intensity amplitude can be recovered by means of a subsequent increase in the substrate temperature. In most depositions of Fe layers the first 5–8 ML were grown at RT. The substrate temperature was then increased to 150–240 °C. The Fe layers deposited in this manner can be grown in a quasilayer by layer manner in which the growth proceeds by terrace nucleation and subsequent attachment of adatoms to the atomic steps of the newly formed atomic islands. The surface roughness of the Fe films was mostly confined to the top two atomic layers.[7]

All samples prior to their removal from the vacuum system were covered by a 20-ML-thick epitaxial Au(001) layer. The growth of Au exhibited well defined RHEED intensity oscillations and the surface was terminated by a 5×1 reconstruction typical for Au(001).

MAGNETIC STUDIES

Quantitative Brillouin light-scattering (BLS) studies[4,8] have clearly exhibited short-wavelength oscillations in the exchange coupling; see Fig. 4. These studies showed also that the exchange coupling through Cr(001) contains both oscillatory bilinear J_1 and positive biquadratic J_2 exchange coupling terms. The exchange energy can be written

$$E=-J_1\cos(\theta)+J_2\cos^2(\theta), \tag{1}$$

where θ is the angle between magnetic moments of the Fe film and the surface magnetization of the Fe whisker.

The results of the exchange coupling measurements can be described as follows. The exchange coupling crosses to antiferromagnetic coupling at 4 ML of Cr. For Cr spacer thicknesses $d_{\mathrm{Cr}}<8$ ML the strength of the short-wavelength oscillations is quite weak, ~0.1 ergs/cm^2. The exchange coupling in this range is antiferromagnetic only due to the presence of an antiferromagnetic (AF) long-wavelength bias. This AF bias is peaked around 6–7 ML. It is interesting to note that the strength of the long-wavelength AF bias is very nearly the same as that observed in Fe/Cr/Fe(001) epitaxial multilayers prepared by sputtering where the relatively large interface roughness annihilated the presence of the short-wavelength oscillations.[9] The exchange coupling in these sputtered films showed long-wavelength oscillations with a rapidly decreasing strength of the coupling for thicknesses greater than 10 ML of Cr. It follows that the antiferromagnetic bias shown in Fig. 4 for the first 7–8 ML of Cr is most likely due to the long-wavelength oscillations in the exchange coupling. For a Cr spacer thicker than 8 ML, $d_{\mathrm{Cr}}>8$ ML, the exchange coupling is dominated by the short-wavelength oscillations. In this thickness range the samples are antiferromagnetically (AF) coupled for an odd number of Cr atomic layers, $J_{\mathrm{tot}}=|J_1-2J_2|\sim1.0-1.5$ ergs/cm^2, and ferromagnetically (FM) coupled for an even number of Cr atomic layers, $J_{\mathrm{tot}}\sim0.2$ ergs/cm^2. The total exchange coupling through the Cr spacer for the parallel orientation of magnetic moments is given by $J_{\mathrm{tot}}=J_1-2J_2$, and therefore the biquadratic exchange contribution $J_2>0$ increases the strength of the AF coupling, and decreases the strength of the FM coupling. The biquadratic coupling J_2 was found to be approximately 0.25 ergs/cm^2, nearly independent of Cr film thickness. The observed weak FM coupling implies that $J_1\sim0.2+2J_2$ and leads to a bilinear exchange coupling strength of $J_1=0.7$ ergs/cm^2 for FM coupled samples.

The coupling between the Fe and Cr atoms at the Fe/Cr interface is expected to be strongly antiferromagnetic[10,11] and in consequence the spin-density wave in Cr is locked to the orientation of the Fe magnetic moments. Since the period of the short-wavelength oscillations is close to 2 ML one would expect AF coupling for an even number of Cr atomic layers and FM coupling for an odd number of Cr atomic layers. For the period $\lambda=2.11$ ML the first phase slip in the short-wavelength coupling is predicted to occur at 20 ML. Surprisingly the SEMPA (Ref. 12) and BLS (Ref. 13) measurements showed clearly that the phase of the short-wavelength oscillations is exactly opposite to that expected. It is also important to note that the strength of the exchange coupling $|J_{\mathrm{max}}|\sim1.$ ergs/cm^2 was found to be much less than that obtained from the first-principles calculations, $J_1=30$ ergs/cm^2.[14] Our studies showed that the strength of the bilinear exchange coupling J_1 is very sensitive to the initial growth conditions: a lower initial substrate temperature results in a larger exchange coupling strength. The bilinear exchange coupling can be changed by as much as a factor of 5 by varying the substrate temperature during the growth of the first Cr atomic layer.[15] This behavior led us to believe that the atomic formation of the Cr layer is more complex than had been previously acknowledged. Angular resolved Auger spectroscopy (ARAES),[15–17] STM,[18] and proton induced Auger electron spectroscopy (AES) (Ref. 19) have shown that the formation of the Fe/Cr(001) interface is strongly affected by an interface atom exchange mechanism (interface alloying). The above studies revealed very clearly that the Cr undergoes interface mixing when the substrate temperature is adjusted for optimum growth. The tunneling spectroscopy measurements suggested that the surface is predominantly Cr at a Cr coverage of ~2 ML.[18] The ARAES showed that the interface mixing was confined mainly to two Fe atomic layers; see Fig. 5. The proton induced AES studies[19] also showed that the interface atom exchange process does not proceed appreciably for layers beyond the Fe/Cr interface. The ARAES studies showed that interface alloying during the growth already starts at low substrate temperatures, $T_{\mathrm{sub}}\sim100$ °C. The interface alloying increases with increasing substrate temperature; see Fig. 5. It should be noted that interface alloying due to the atom exchange mechanism is not, in general, symmetric: it occurs chiefly at one interface.[7] Interface alloying is driven by the difference in binding energies between the substrate and adatoms. The binding energies are proportional to the melting points of the solids. Interface alloying has been observed in systems for which the substrates have lower melting points than do the adatom solids.[7,20] The melting point of Fe (1808 K) is lower than the melting point of Cr (2130 K) and thus the condition for interface alloying at the Fe/Cr interface is satisfied, but the condition is not satisfied at the Cr/Fe interface. Mössbauer studies by Keune and Schrör *et al.*[21] have shown that interface mixing for the deposition of Fe atoms on a Cr sub-

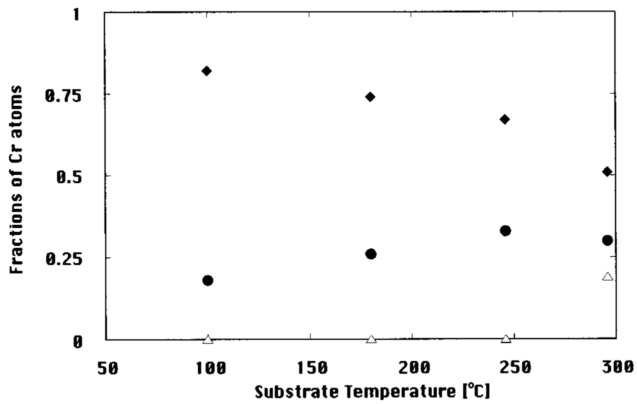

FIG. 5. The substrate temperature dependence of the fraction of Cr atoms in the first layer deposited on an Fe-whisker substrate (♦), the fraction of Cr atoms contained in the whisker surface layer (●), and the fraction of Cr atoms contained in the first whisker subsurface layer (△). The fractional coverages were obtained from fitting the angular dependence of the Auger Cr (529 eV) peak intensity [using angular resolved Auger electron spectroscopy (ARAES)] with the SSC computer program provided by Chuck Fadley.[50]

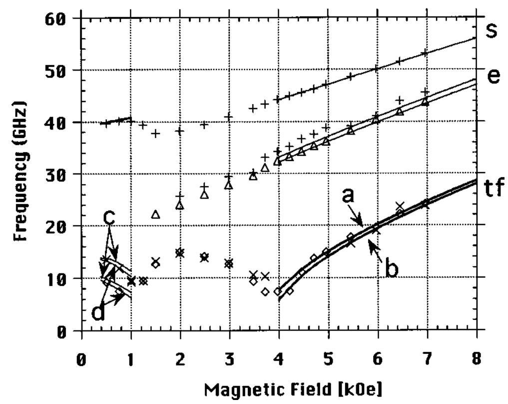

FIG. 6. A comparison of observed and calculated scattered light frequency shifts for a structure composed of a bulk whisker-Fe(001) substrate, an 11-ML-thick Cr(001) spacer, a 20-ML-thick Fe(001) film, and a 20-ML-thick Au cover layer. The 5145 Å laser light was incident at 45° and the scattered light was collected in the backscattering configuration. The saturation magnetization was taken to be 21.4 kOe for both the bulk Fe and the Fe thin film. The in-plane cubic anisotropies used were 4.76×10^5 erg/cm^3 for the bulk Fe and 3.5×10^5 erg/cm^3 for the thin film. The perpendicular uniaxial surface anisotropies were taken to be 0.5 erg/cm^2 for the bulk Fe and 1.0 erg/cm^2 for the Fe thin film. The uniaxial surface anisotropy for the Fe film was obtained by independent measurements using Ag substrate/Cr/Fe/Au(001) samples. The in-plane cubic anisotropy for the Fe film was determined using Eq. (1.40) in Ref. 4. (+) Observed surface mode frequencies *s* and up-shifted bulk magnon edge frequencies *e*. (△) Observed down-shifted bulk edge frequencies *e*. (◇) Observed up-shifted thin-film frequencies *tf*. (×) Observed down-shifted thin-film frequencies *tf*. Curve (a) has been calculated using a coupling strength $J_{tot}=J_1-2J_2=-1.45$ ergs/cm^2; curve (b) has been calculated for $J_{tot}=-1.50$ ergs/cm^2; curve (c) has been calculated using $J_{tot}=J_1+2J_2=-0.70$ ergs/cm^2; curve (d) has been calculated using $J_{tot}=-0.60$ ergs/cm^2. The cusp fields are $H_1=1.0$ kOe and $H_2=4.0$ kOe. We estimate that $J_1=-1.1$ erg/cm^2, and $J_2=0.2$ erg/cm^2.

strate is not important for deposition temperatures near room temperature such as we used to deposit Fe films on the Cr layers.

The results of typical BLS and MOKE studies are shown in Figs. 6 and 7. The MOKE and BLS measurements exhibit two critical fields. For fields $H>H_2$ the magnetic moments in the Fe whisker and in the ultrathin film were clearly parallel to the applied external field, and the sample can be fully saturated in sufficiently large fields. For $H<H_2$ the magnetic moments were noncollinear, the magnetic moments deviated from the external field direction. As illustrated in Fig. 6, the Stokes and anti-Stokes modes are split by 4 GHz for fields $H<H_1$ and for a 20-ML-thick Fe film. This splitting is the consequence of dipolar coupling when the direction of the dc magnetic moments in the Fe whisker and in the Fe film are antiparallel.[8] In all our samples the field dependence of the magnetization loops is consistent with the assumption that the angular variation of the exchange coupling can be expressed in terms of bilinear and biquadratic exchange coupling terms [Eq. (1)]. The separation between the H_2 and H_1 fields, $\Delta=H_2-H_1$, calculated using only the bilinear exchange coupling term was always smaller than the measured separation Δ_m: one needed to add the biquadratic exchange coupling term with J_2 positive to obtain the observed separation Δ_m. The positions of critical fields H_1 and H_2 were calculated using a full micromagnetic calculation for an Fe-whisker/Cr/20 Fe sample including both bilinear and biquadratic exchange coupling terms. In these calculations the angular spatial variations of the magnetic moments inside the whisker and across the film were included.[22] The theory described in Ref. 22 was used to determine the strengths of J_1 and J_2 by comparison with the experimental Kerr data. A large number of measurements showed that a part of J_2 was proportional to the measured value of J_1. We found that $J_2\sim0.1+(0.16|J_1|)$. This is an interesting result that requires a brief comment. Stoeffler and Gautier predicted the presence of a biquadratic exchange coupling term for Fe layers coupled through Cr(001).[14] The origin of this coupling is the complex behavior of the Cr spin-density wave when the magnetic moments of the ferromagnetic layers are not locked in phase with the spin-density wave of the Cr spacer in the lowest energy state. J_2 is expected to be ~14% of $|J_1|$. The coefficient 0.16 can indicate that a part of J_2 is related to the intrinsic contribution of the biquadratic exchange coupling in spin-density wave Cr. Biquadratic exchange coupling in spacers that possess short-wavelength oscillations in J_1 can also be caused by lateral variations of the spacer thickness.[23,24] The strength of the biquadratic exchange coupling depends on the lateral scale of the thickness variations. When the lateral thickness variations occur on a scale that is much less than the in-plane exchange correlation length, that is on a length of the order of the domain-wall width, the biquadratic exchange coupling due to lateral thickness variations becomes negligible; see Eq. (2.17) in Ref. 4. The STM studies[18] have shown that the interface alloying is distributed laterally on an atomic length scale, and in this case the interface roughness caused by any interface alloying should make a negligible contribution to J_2. The atomic roughness at the second interface, Cr/Fe, is different from that at the first interface, Fe/Cr, and is not due to interface alloying. The roughness at the second interface can be caused by two ef-

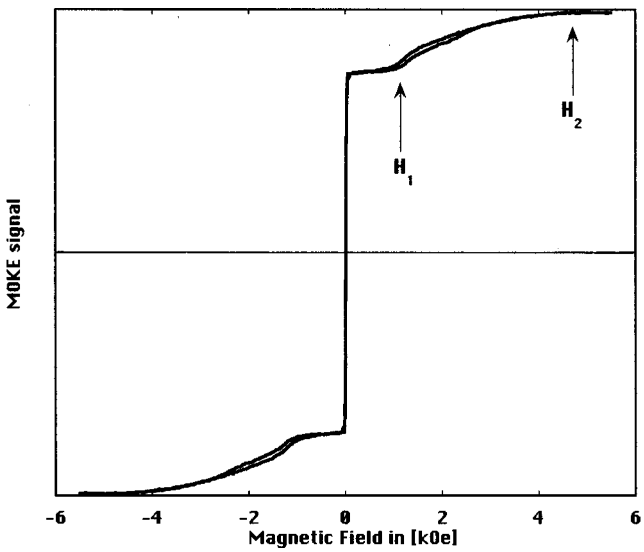

FIG. 7. The longitudinal MOKE signal for the same sample as in Fig. 6 as a function of the applied magnetic field. $H_1 = 1.3$ kOe and $H_2 = 4.8$ kOe. Notice that the saturation field H_2 is higher in MOKE than that in BLS. The difference between the MOKE and BLS measurements of the critical field H_2 is caused by lateral inhomogeneities in exchange coupling, see the text for the details. These values of H_1 and H_2 correspond to $J_1 = -1.16$ erg/cm^2 and $J_2 = 0.27$ erg/cm^2.

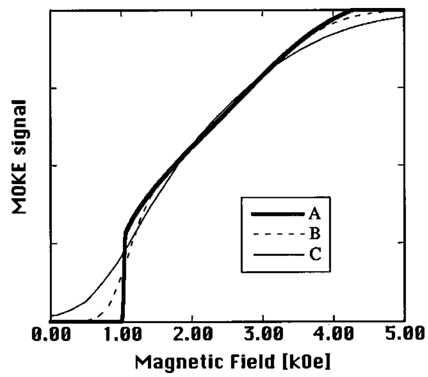

FIG. 8. Calculated MOKE signal for a 20 ML Fe(001) film exchange coupled to a bulk Fe(001) substrate and assuming an inhomogeneous distribution of the bilinear coupling strength J_1. The biquadratic coupling strength J_2 has been set equal to 0.3 ergs/cm^2 for all curves. The probability of a coupling strength $J_1 = J, P(J)$, has been taken to be $P(J) = [1/(\pi\Delta J)^{1/2}]^* \exp\{-[(J - \langle J\rangle)/\Delta J]^2\}$ with $\langle J\rangle = -1$ erg/cm^2. Curve (A) $\Delta J = 0$. Curve (B) $\Delta J = 0.2$ ergs/cm^2. Curve (C) $\Delta J = 0.5$ ergs/cm^2.

fects: (a) by terminating the Cr growth not exactly at a full coverage of the last atomic layer, and (b) by changing the distribution of atomic terraces during the growth. Effect (a) most likely plays a minor role in carefully prepared samples where the Cr interlayer is continuously monitored by means of RHEED; see above. The interface roughness due to the redistribution of atomic steps at the top Cr surface, case (b), could lead to the most serious consequences. In this case the Cr thickness varies by 1 ML across the sample surface and the lateral scale of the variations in Cr thickness would be given by the mean distance between the Cr nucleation centers, ~800–900 Å. This thickness variation could result in a significant decrease of the bilinear exchange coupling J_1 due to a near cancellation of positive and negative contributions to J_1. At the same time the large scale lateral variations in the Cr spacer thickness would lead to a large contribution to J_2. For such large scale lateral inhomogeneities the approximations used to derive Slonczewski's formula[23] are not directly applicable. The angle between the magnetic moments of the two coupled Fe layers deviates strongly from its mean value from place to place along the surface. For large lateral inhomogeneities micromagnetic calculations were carried out by Arrott.[24] Model calculations showed that the calculated magnetization loops were similar to those obtained using the bilinear and biquadratic exchange interactions, Eq. (1), with a strong biquadratic exchange coupling contribution for which the total magnetic moment does not show any lower critical field H_1, and the ground state is noncollinear. However, the measured hysteresis loops and BLS measurements exhibit a behavior in which the biquadratic exchange coupling is much less than J_1. Therefore one can conclude that the observed low values of the bilinear exchange coupling are not due to the redistribution of atomic steps, but are more likely to be the direct consequence of interface alloying.

Neutron-diffraction and MOKE studies by the Bochum group,[25] using Fe/Cr/Fe(001) samples having a high density of atomic steps, showed that the ground state in Fe/Cr/Fe(001) multilayers exhibits a noncollinear orientation of the magnetic moments for which the approach to saturation can be described by the Slonczewski magnetic proximity model[26] in which the exchange energy depends quadratically on the angle between the magnetic moments of the ferromagnetic layers, $\sim(\Delta\theta)^2$. All our MOKE and BLS measurements carried out on Fe-whisker/Cr/Fe(001) samples that were prepared with the best possible interfaces (low density of atomic steps) are consistent with the use of bilinear and biquadratic exchange coupling terms, Eq. (1). In particular, the BLS data show unambiguously that for fields slightly above the critical field H_2 the iron film and whisker moments are parallel, whereas for fields slightly less than the critical field H_1 the thin film and whisker moments are antiparallel.[8] The approach to saturation at the critical field H_2 as observed using MOKE, and the onset of the antiferromagnetic configuration at the critical field H_1, see Fig. 7, is more gradual than is calculated using the sum of bilinear and biquadratic exchange coupling terms of Eq. (1); see Fig. 8, curve A. The calculated approach to saturation clearly exhibits a well defined kink at the field H_2: the experimental measurements usually show a concave gradual approach to saturation; see Fig. 7. According to the calculations the antiferromagnetic configuration of the Fe magnetic moments is reached via a first-order phase jump; the experimental measurements show a more gradual s-shaped change. These experimental MOKE features can be explained by an inhomogeneous distribution of the exchange coupling strength. A 10% variation in the exchange coupling across the measured area would result in hysteresis loops that are very similar to those observed using MOKE; compare Fig. 7 with Fig. 8. In fact an inhomogeneous distribution of the exchange coupling also explains observed differences between values of the critical fields H_2 that have been obtained using the MOKE and the BLS techniques. The BLS measurements on Fe/Cr/Fe(001) always yield lower values for the critical field H_2, with corresponding lower values of the exchange coupling strength, compared with that obtained using MOKE. The BLS thin-film resonant modes were also visibly broadened for external fields greater than the saturation field H_2, where the thin-film magnetic moment is parallel to the Fe-whisker moment. (The thin-film resonance mode was observed to be much narrower at a field midway between H_1 and H_2 where the thin-film and whisker magnetizations are nearly orthogonal.[8]) The difference between critical fields measured using BLS and MOKE, as well as the broadening of the BLS signal for fields greater than H_2, can be explained by an inhomogeneous distribution of J_1 and J_2. The BLS technique measures the frequencies of the rf resonance modes. The mode corresponding to the thin Fe film covering the Cr spacer exhibits a resonance at a field that is the algebraic average of local inhomogeneous resonance fields (corresponding to the distribution of local exchange coupling strengths), and the broadening of this BLS resonance peak is related to the distribution of the local resonance fields. On the other hand, in the MOKE studies a complete saturation is observed only after the external field reaches a value corresponding to the maximum value of the critical field H_2 distribution: this maximum value of H_2 is larger than the field corresponding to the rf resonance peak (algebraic average). The observed differences between the MOKE and BLS measurements clearly indicate that the coupling through Cr is not even homogeneous across an area a few micrometers in diameter corresponding to the laser spot size in the BLS measurements. It is important to point out that it is very difficult to exactly reproduce the strength of the exchange coupling from one Cr growth to the next. Small variations in the growth conditions can lead to variations in J_1 as large as a factor of two. It was possible to obtain the results shown in Fig. 4 only after an extensive series of experiments using different growth conditions. Quantitatively reproducible results were obtained only for a very specific set of conditions: the RHEED intensity oscillations must be cuspy and unattenuated during the growth; the RHEED specular spot profile corresponding to an intensity maximum must be as narrow as the spot profile for the bare whisker surface; and, most importantly, the minimum RHEED intensities must remain very close to zero throughout the entire growth. It is difficult to satisfy all of these conditions during any one growth. The RHEED intensity oscillations shown in Fig. 2 in which the intensity minima gradually increase during the growth is common; see Fig. 2 caption. The problem in establishing the required growth conditions lies in the fact that the Fe whiskers must be lightly attached to the Mo support block in order to avoid damaging them. Thus the thermal contact between whisker and support block is not reproducible, and hence the temperature of the whisker cannot be precisely measured. Variations in whisker temperature during the Cr growth lead to variations in interface alloying at the Fe/Cr interface, and hence lead to variations in the coupling strength between the Fe whisker and a subsequently grown Fe film.

In our view the observation of a gradual approach to saturation using MOKE does not necessarily imply an exchange coupling energy proportional to $(\Delta\theta)^2$ as proposed in the Slonczewski magnetic proximity model.[26] MOKE measurements can only be carried out to fields as large as the maximum available. This means that if sufficiently large applied fields are not available the approach to saturation in a strongly coupled but inhomogeneous system can easily be interpreted in terms of the Slonczewski magnetic proximity model. One should always consider the possibility of inhomogeneous coupling. The combination of BLS and MOKE measurements provides a test for the presence of inhomogeneous interlayer coupling.

Recent calculations[27] predict that the exchange coupling through Cr(001) spacers which have ideal interfaces can be described by Slonczewski's proximity model[26] in which the exchange energy increases quadratically with the angle between the magnetic moments of the Fe layers, exchange energy $\sim(\Delta\Theta)^2$. In that case, for AF coupling, the total magnetic moment approaches saturation gradually; there is no torque free solution in high fields. The BLS and MOKE measurements using Fe-whisker/Cr/Fe(001) samples having a low density of atomic steps exhibit much weaker coupling than that obtained from the first-principles calculations,[27] and there is strong evidence (see above) that the measured samples can be fully saturated in sufficiently large external fields. The discrepancy between the experimental results and the theoretical expectations is very likely caused by the presence of interface alloying at the Fe/Cr interface. Interface alloying significantly decreases the exchange coupling and may even change the functional angular dependence of the coupling between the Fe layer magnetic moments. It is interesting to note that the first calculations carried out by Stoeffler and Gautier[14] yielded an angular dependence that is well described by bilinear and biquadratic exchange coupling terms [Eq. (1)] with $J_2/J_1\sim14\%$. This calculation is in agreement with our observations. However, the calculations were carried out for a spin system that was not completely relaxed. Fully relaxed first-principle calculations of the angular dependence of the interlayer exchange coupling in samples having an intermixed Fe/Cr interface are needed in order to address this problem of the theoretical angular dependence of the exchange coupling in the presence of alloyed interfaces.

ROLE OF INTERFACE ALLOYING

Recently, Freyss, Stoeffler, and Dreyssé[28] investigated the phase of the exchange coupling for intermixed Fe/Cr interfaces. The calculations were carried out using a tight-binding *d*-band Hamiltonian and a real-space recursive method for two mixed layers: Fe(001)/Cr_xFe_{1-x}/$Cr_{1-x}Fe_x$/Cr_n, where *n* represents the number of pure Cr atomic layers. This simulates our experimental studies which were carried out on specimens for which the first few atomic Cr layers were grown at lower substrate temperatures where the surface alloying is mainly confined to the two interface atomic layers. The calculations were able to account for two important experimental observations. First, the crossover to antiferromagnetic coupling and onset of short-wavelength oscillations was predicted to occur at 4–5 ML of Cr, in good agreement with our observations, see Fig. 4, and in agreement with the NIST studies using the SEMPA imaging technique. Second, the phase expected for perfect interfaces (AF coupling for an even number of Cr monolayers, FM coupling for an odd number of Cr monolayers) was found to be reversed for $x\geqslant0.2$. In other words, a Cr layer containing more than 20%

iron acted as if it were part of the ferromagnetic iron layer rather than acting like part of the Cr spacer layer. This result is also in very good agreement with our studies. Samples for which the Fe/Cr interface was prepared at 150 °C, and showing only a weak interface diffusion ($x \sim 0.2$),[15,16] exhibited a phase for the exchange coupling that was reversed from that expected for perfect interfaces.

We tried to avoid interface mixing by decreasing the substrate temperature during the growth of the first Cr atomic layer. We found that the quality of the subsequently deposited Cr layers was noticeably poorer once the initial substrate temperature was decreased below 100 °C. Since we were not able to defeat interface mixing using a direct approach, we decided to use heterogeneous Cr spacers. N Cr layers were deposited on the Fe-whisker substrate using the standard recipe known to produce a smooth layer-by-layer growth ($T_{s,\mathrm{opt}} \sim 300\,°\mathrm{C}$, see the section on growth). However, the last deposited atomic layer, the $N+1$ layer, was prepared using codeposition of Cr with Fe to produce a Cr-Fe alloy. RHEED intensity oscillations and RHEED patterns were basically unchanged and showed that the Cr-Fe alloy layer was atomically flat. Two alloy concentrations were used: Cr 85%-Fe 15% and Cr 65%-Fe 35%. BLS and MOKE measurements[29] revealed that the exchange coupling between a thin (20 ML) iron film and the whisker substrate was essentially the same as that observed for a pure N layer Cr spacer layer. The system behaved as if the Cr alloy layer formed part of the iron film. This result strongly supports the idea that interface alloying at the Fe-whisker/Cr interface leads to the observed phase reversal of the short-wavelength exchange coupling oscillations relative to a system having perfect interfaces. Moreover, this picture has recently received strong support from magnetic circular dichroism measurements carried out by Schneider *et al.*[30] Their data have shown that the magnetic moment of the first Cr layer deposited on iron is parallel with the iron moment.

The fact that one needs only a small concentration of Fe to reverse the phase of the coupling is a rather surprising result, especially considering the further recent calculations by the Strasbourg group.[28] The article by Freyss, Stoeffler, and Dreyssé mentioned above[28] also contains calculations carried out for a single interfacial alloyed layer: they treated the system $\mathrm{Cr}_n/\mathrm{Cr}_x\mathrm{Fe}_{1-x}/\mathrm{Fe}(001)$, where Fe(001) is a thick substrate and n is the number of pure Cr atomic layers. Their calculations showed that for iron concentrations less than 50% the Cr-alloy layer behaves like a pure Cr atomic layer. This result is not in agreement with our experimental observations that even a concentration of Fe as small as 15% causes the alloy layer to behave like Fe rather than like pure Cr. The discrepancy with theory remains to be explained.

ROLE OF MULTIPLE SCATTERING

Heterogeneous Cr spacers were prepared in order to test the effect of interface composition on the exchange coupling strength. Two specimens were grown with a Cu interface layer between the Cr spacer and the Fe thin film: Fe-whisker/11Cr/1Cu/20Fe(001)/20Au and Fe-whisker/11Cr/2Cu/20Fe(001)/20Au, where the integers represent the number of atomic layers. Two specimens were grown with a silver interface layer: Fe-whisker/11Cr/1Ag/20Fe(001)/20Au and Fe-whisker/11Cr/2Ag/20Fe(001)/20Au. The growth of Ag at $T_{\mathrm{sub}}=105\,°\mathrm{C}$ resulted in nearly perfect layer by layer growth. The growth of Cu using $T_{\mathrm{sub}}=65\,°\mathrm{C}$ was less perfect but still showed well defined RHEED oscillations, even for the second anti-Bragg condition, indicating that the atomic deposition of the Cu layers was reasonably smooth. Measurements carried out using BLS and MOKE gave similar results, and the results for a 1 ML interface layer were similar to the results for the 2 ML interface layer for both the Cu and Ag layers. The results of BLS and MOKE studies were qualitatively the same as those shown in Figs. 6 and 7. Specimens were prepared during the same molecular beam epitaxy (MBE) growth having two different spacer configurations on the same whisker. The Cr(001) spacer was common to both regions, but a shutter covered half the whisker during the Cu and Ag depositions. In this way the effect of Cu or Ag layers on the exchange coupling could be studied free from uncertainties in the coupling strength due to slight variations in the Cr growth conditions. The results of the measurements using 2 ML of Cu or Ag were as follows:

(1a) Fe whisker/11Cr/20Fe/20Au; $J_1=-0.4\ \mathrm{erg/cm^2}$, $J_2=0.18\ \mathrm{erg/cm^2}$;

(1b) Fe whisker/11Cr/2Cu/20Fe/20Au; $J_1=-0.81\ \mathrm{erg/cm^2}$, $J_2=0.3\ \mathrm{erg/cm^2}$;

(2a) Fe whisker/11Cr/20Fe/20Au; $J_1=-1.3\ \mathrm{erg/cm^2}$, $J_2=0.35\ \mathrm{erg/cm^2}$;

(2b) Fe whisker/11Cr/2Ag/20Fe/20Au; $J_1=-1.0\ \mathrm{erg/cm^2}$, $J_2=0.35\ \mathrm{erg/cm^2}$.

The behavior of the exchange coupling in the Fe-whisker/11Cr/1–2Cu/Fe(001) samples is most surprising. The strength of the exchange coupling in these samples was found to increase twofold compared to that observed in samples having a simple 11 ML Cr spacer layer. This is an unexpected result. In all of our previous studies, using Fe/Cu/Fe(001) structures containing a wide range of heterogeneous Cu spacers, the exchange coupling was always found to decrease due to the presence of alloyed atomic layers inside the nonmagnetic spacer.[31] The situation for the Fe/Cr/Cu/Fe(001) specimens is definitely different. Mirbt and Johansson presented calculations[32] that are in accord with our results. Their calculations show that the enhanced coupling strength in Fe/Cr/Cu/Fe(001) samples is due to a change in the spin dependent reflectivity of the Cr spacer electrons at the Cr/Cu/Fe interface. The presence of the Cu atoms changes the spin dependent interface potential due to hybridization of the Cu electron states with the Fe electron states. Since the Fe majority spin band lies closest to the Fermi level the effect of hybridization will be most pronounced for the majority spin Fe band. The hybridization with Cu results in a downward energy shift that moves the Fe majority spin band below the Fermi level. An energy gap is created at the Cu/Fe interface, and consequently the majority spin electrons in Cr undergo a nearly perfect reflection. The states for minority spin electrons are very little affected by the Cu, and therefore their reflectivity is left unchanged. It follows that the spin reflection asymmetry is increased leading to an increased coupling.[33–35] The effect of a Ag spacer on the coupling in Fe/Cr/Ag/Fe is less dramatic. Calculations show that the spin asymmetry in reflectivity is somewhat decreased leading to an overall decrease in the exchange coupling. The theoretical calculations of Mirbt and Johansson suggested that a proper model for exchange coupling through spin-density waves in Cr has to include two contributions: (a) a spin dependent potential due to the magnetic moments on the antiferromagnetic Cr atoms; (b) a spin dependent potential at the Fe/Cr and Cr/Fe interfaces. The first contribution for Cr layers thinner than 24 ML can be described by a Heisenberg-like Hamiltonian[14] with AF coupling between the Cr magnetic moments on adjacent (001) planes and a strong AF coupling between the Cr and Fe atomic moments at the interfaces for perfect interfaces, and with modified exchange interactions for alloyed interfaces.[28] The second contribution leads to spin dependent reflectivities at the interfaces. The spin reflectivites are parameters closely associated with paramagnetic behavior. In the Mirbt and Johansson calculations[32] the contribution of the spin-density wave oscillatory exchange coupling is out of phase with that of the multiple scattering in samples of Fe/Cr/Cu/Fe. This means that according to their theory the strength of the exchange coupling should be decreased by the presence of a Cu layer at the interface. However, the measured exchange coupling in Fe/Cr/Cu/Fe(001) samples was found to be increased compared to that in samples having a pure Cr spacer layer (Fe/Cr/Fe). The experimental result implies that the spin-density and multiple scattering contributions to the total exchange coupling act in phase. The phase of the multiple scattering is very dependent on the structural details of the interfaces, consequently it is not surprising that the observed phase of the multiple scattering in real Fe/Cr/Fe samples was found to be opposite to that calculated assuming ideally smooth interfaces.

MAGNETIC STATE OF Mn(001) IN Fe/Cr/Mn/Fe

It is known that even small concentrations of Mn in Cr results in a strong and commensurate antiferromagnetism.[36] It was therefore thought to be of interest to investigate the phase and the strength of the exchange coupling between Fe layers separated by layers of a Cr-Mn alloy. To this end we attempted to grow Fe/Cr-Mn/Fe structures. Unfortunately we found that the Mn atoms have very strong tendency to segregate on the surface during the growth. It was necessary to maintain a substrate temperature greater than 200 °C in order to obtain a good layer by layer growth. At this temperature the top surface layer contained a strongly enhanced concentration of Mn (~50%). In view of this surface segragation, and since the interfaces play a very crucial role in exchange coupling, we decided to grow pure layers of Mn between the Cr and the Fe layers. Eleven or 12 ML of Cr were grown on an Fe(001) whisker using growth conditions optimized for layer-by-layer growth (see the section on growth). Mn layers were deposited on the Cr at a substrate temperature of 120 °C. The substrate was allowed to cool to room temperatures and 20 ML of Fe(001) were deposited on the Mn, and a protective layer of 20 ML of Au(001) were deposited on the iron. At 120 °C the deposition of the first two atomic layers of Mn proceeds in a good layer by layer growth with large RHEED intensity oscillations at the second anti-Bragg scattering condition. At substrate temperatures well below 100 °C the Mn does not segregate on Fe. In this way one is able to grow well defined Fe/Cr/Mn/Fe structures having smooth and abrupt interfaces. It should be pointed out that the top Cr surface atomic layer is smooth, with large atomic terraces corresponding to those of the Fe whisker, and is unaffected by interface alloying during the deposition of the Mn. Therefore variations in the exchange coupling due to the addition of the Mn layers are primarily due to the presence of the Mn atomic layers and their magnetic state. There is no intermediate Cr-Mn mixed region similar to the Cr-Fe mixed region that occurs at the Fe-whisker/Cr interface.

The following samples were studied: (i) Fe(001)/11Cr/1Mn/20Fe(001)/20Au; (ii) Fe(001)/11Cr/2Mn/20Fe(001)/20Au; (iii) Fe(001)/12Cr/3Mn/20Fe(001)/20Au; and (iv) Fe(001)/11Cr/3Mn/20Fe(001)/20Au. Sample (i) was grown together with Fe(001)/11Cr/20Fe(001)/20Au on the same whisker using a shutter. This configuration allowed one to compare the strength of the coupling in samples with and without Mn but having a common Cr layer. MOKE and BLS measurements showed that 1 ML of Mn did not change the phase of the coupling. The coupling was found to be AF for both the 11 ML Cr spacer and the composite spacer, specimen (i). The BLS and MOKE results were qualitatively similar to those shown in Figs. 6 and 7. The exchange coupling strengths for (i) were as follows: Fe(001)/11Cr/20Fe/20Au; $J_1=-0.68\,\mathrm{erg/cm^2}$, $J_2=0.24\,\mathrm{erg/cm^2}$; Fe(001)/11Cr/1Mn/20Fe/20Au; $J_1=-1.7\,\mathrm{erg/cm^2}$, $J_2=0.55\,\mathrm{erg/cm^2}$. Note that the exchange coupling was significantly enhanced by the Mn layer. It is commonly believed that the Mn magnetic moment is strongly ferromagnetically coupled to the Fe magnetic moment[37,38] and therefore the sign of the coupling should be unchanged in agreement with the observations. For the composite specimen containing 2 ML of Mn, Fe(001)/11Cr/2Mn/20Fe/20Au, the results were $J_1=-0.62\,\mathrm{erg/cm^2}$, $J_2=0.14\,\mathrm{erg/cm^2}$. Therefore the phase of the coupling was again found to be the same as that for the pure Cr layer. The second atomic layer of Mn was expected to be AF aligned with respect to the first Mn atomic layer.[37,38] Assuming that the Mn(001) planes are magnetically uncompensated, this would lead to a phase reversal of the exchange coupling when specimen (ii) was compared with the specimen containing a pure Cr interlayer. Such a phase reversal was not observed. The exchange coupling in sample (iii) was found to be ferromagnetic and the coupling in sample (iv) was found to be antiferromagnetic. This shows that the exchange coupling oscillates with the same phase and the same periodicity of 2 ML as was observed using pure Cr spacer layers.

Although the sign of the coupling for the 3 ML Mn case [sample (iv)] was the same as for a pure Cr spacer of the same thickness, the magnetizations of the whisker and the 20 ML Fe film were noncollinear in the ground state, $H=0$; see Fig. 9. For an 11 ML pure Cr spacer the whisker and Fe thin film magnetizations are oriented antiparallel at $H=0$.

The presence of a strong biquadratic exchange coupling in the sample with the 3-ML-thick Mn layer, sample (iv), can be related to the interface roughness. The growth of the third atomic layer of Mn did not proceed as well as for the first two atomic layers. Consequently the third deposited Mn atomic layer was probably partially filled, resulting in atomic terraces and corresponding atomic steps. The magnetic moments around the atomic steps are probably magnetically uncompensated, and this could result in a random oscillation of the magnetization between being parallel and antiparallel to the Fe film magnetic moment. The coupling between the Fe

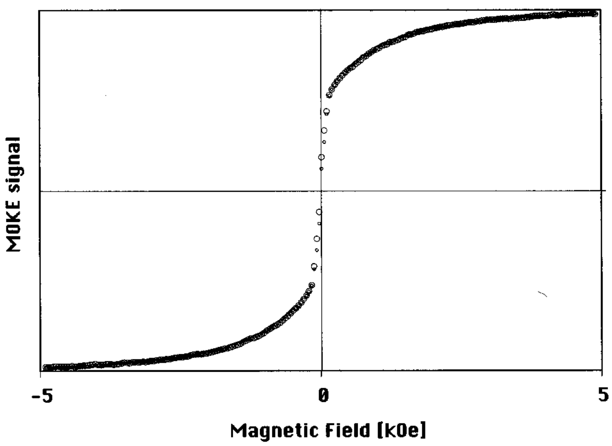

FIG. 9. Longitudinal MOKE signal for the sample Fe/11Cr/3Mn/20Fe/20Au(001). $J_2 \sim 0.8\,\mathrm{erg/cm^2}$, $J_1 < J_2$. The rapid change in the MOKE signal around $H=0$ corresponds to the remagnetization process of the Fe whisker. Note that there is no field region where the magnetic moment of the 20-ML-thick Fe film lies antiparallel to the Fe-whisker magnetization. The ground state is noncollinear.

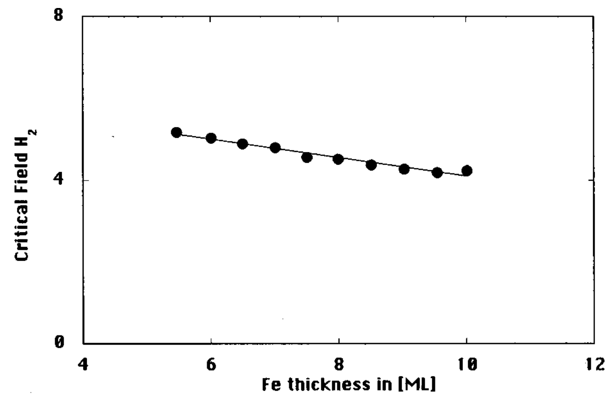

FIG. 10. The critical field H_2 obtained from MOKE measurements as a function of the Fe(001) thickness for an Fe-whisker/11Cr/wedged Fe/20Au specimen. The Cr layer was deposited using optimal conditions for a smooth growth. The wedge-shaped iron film was grown by means of a slowly moving shutter.

and Mn atoms is most likely confined to a nearest neighbor exchange interaction, and that would result in a lateral spatial variation of the coupling between the Mn and Fe atomic moments thereby providing the proper conditions for the onset of a strong biquadratic exchange coupling.[23]

The assumption that the magnetic state of Mn in Fe/Cr/Mn/Fe(001) structures can be described by commensurate antiferromagnetism with uncompensated (001) planes is not necessarily correct. In recent calculations by Krüger *et al.*[39] the magnetic structure of bct Mn in bulk was studied as a function of the tetragonal distortion, c/a. The calculations showed that bct Mn grown on Fe(001) is in a magnetic state that is at the border line between the AF1 configuration, having the magnetic moments parallel in (001) planes, and the AF3(110) configuration, having ferromagnetic planes oriented along (110), and fully compensated (001) planes having zero net magnetic moment. The calculations showed that the lowest energy state is just marginally the AF3(110) state, only 4 meV/atom lower in energy than the AF1 state. The AF3(110) state would not lead to an alternating sign of exchange coupling with increasing Mn thickness, and would be in agreement with our experimental observations. The above calculations show that the independence of the sign of the exchange coupling on Mn thickness is not at variance with expectations based on the assumption that Mn grown on Fe(001) takes on the bulk bct Mn structure. This view is further supported by recent theoretical and experimental studies. Wu and Freeman[40] showed that one atomic layer of Mn on Fe(001) is compensated (ordered antiferromagnetically). Recent experimental studies using magnetic circular x-ray dichroism (MCXD) (Refs. 41 and 42) showed no net magnetic moment for higher Mn coverages on Fe(001). Also, recent electron capture experiments using the scattering of He^+ ions showed no magnetic moment for higher Mn coverages on Fe.[43] These results are consistent with the conclusion that the Mn(001) atomic planes are antiferromagnetically ordered and magnetically compensated.

THE DEPENDENCE OF THE EXCHANGE COUPLING ON THE THICKNESS OF THE Fe FILM

The spin-dependent potential in multilayer films creates electron confinement and resonant states which are responsible for the oscillatory behavior of the exchange coupling. According to theoretical calculations[44,45] such states are not restricted to nonmagnetic spacers, but are also present inside the ferromagnetic layers, and the coupling cannot be entirely described by an interaction that is localized at the interfaces. The energy terms coming from the electron confinement in the ferromagent layers due to multiple reflections and the interference of such states with the states inside the spacer layer results in a variation of the interlayer exchange coupling with the ferromagnetic layer thickness. Calculations and experimental studies on the Co/Cu/Co(001) (Ref. 46) and the Fe/Au/Fe(001) systems[47] have shown that the exchange coupling contains a component that oscillates as a function of the ferromagnetic layer thickness. However, the oscillatory part is smaller than the total strength of the exchange coupling so that the sign of the coupling is determined by the thickness of the nonferromagnetic spacer layer. Okuno and Inomata[48] reported a strong oscillatory dependence on iron thickness of the exchange coupling in Fe/Cr/Fe(001) specimens. The period of the oscillation was 6 ML. However, the specimens used in their work were multilayers characterized by rough interfaces, and the exchange coupling exhibited no short period (2 ML) variations with Cr thickness. We have measured the dependence of the exchange coupling strength on iron film thickness using an Fe(001) whisker substrate, an 11 ML layer of Cr, and a wedge-shaped Fe layer prepared by means of a slowly moving shutter. This structure was capped by a 20 ML layer of gold. The Cr was deposited using optimum conditions for a smooth growth. The results are shown in Fig. 10. There is no evidence for an oscillatory dependence of the coupling strength on iron film thickness. Figure 10 clearly shows that the upper saturation field H_2 increases gradually with decreasing Fe layer thickness as expected from micromagnetic calculations assuming a constant value for the strength of the interlayer exchange coupling. We have also prepared a specimen Fe(001)/11Cr/nFe/20Au in which four different iron thick-

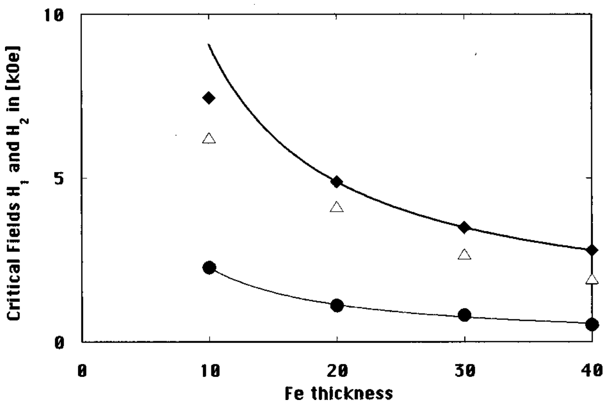

FIG. 11. Fe thickness dependence of the critical field H_2 as measured using MOKE (◆), using BLS (△), and the critical field H_1 obtained from MOKE measurements (●). All four samples were prepared during the same MBE growth. The shutter was moved three times by 2 mm along the whisker during the preparation of the Fe thin films. This means that the Cr(001) spacer was common to all samples and each sample was 2 mm long. The solid lines are computer fits to the experimental data using the functional form (const$+1/d$). This functional form is obtained from simple micromagnetic calculations (Ref. 4) assuming a constant value for the exchange coupling and assuming that the Fe whisker can be treated like a thick film.

nesses corresponding to $n=10$, 20, 30, 40 ML were grown on the same whisker by means of a moveable shutter. Each iron thickness region was 2 mm long. Here again, the Cr layers were deposited using optimum conditions to produce a smooth growth. The results of the measurements are shown in Fig. 11 and listed in Table I. There is no evidence for an oscillatory dependence of the exchange coupling on iron thickness. No dependence on Fe film thickness of the exchange coupling was also reported by Parkin.[49] We conclude that Fe(001)/Cr/Fe(001)/Au samples having a low density of interfacial steps, and that exhibit short-wavelength oscillations as a function of the Cr layer thickness, display no measurable variations of the exchange coupling strength as a function of the Fe film thickness. It appears that the exchange coupling between Fe layers separated by a Cr spacer can be ascribed to interactions that are localized to the interfaces. However, our recent experiments do indicate that the exchange coupling is sensitive to the Fe film thickness when the iron film is capped with Cr to produce the structure Fe(001)/11Cr/Fe/Cr/20Au. Specimens were prepared using two iron whisker substrates; each substrate carried two specimens containing the same 11 ML of Cr. For Fe whisker #1 the structure and coupling strengths observed were: (1a) whisker/11Cr/20Fe/20Au; $J_1=-0.82\ \mathrm{erg/cm^2}$, $J_2=0.3\ \mathrm{erg/cm^2}$; (1b) whisker/11Cr/20Fe/11Cr/20Au; $J_1=-1.6\ \mathrm{erg/cm^2}$, $J_2=0.33\ \mathrm{erg/cm^2}$. Clearly the substitution of Cr for Au at the upper Fe film surface caused a substantial increase in the coupling strength. For Fe whisker #2 the structure and coupling strengths observed were: (2a) whisker/11Cr/18Fe/11Cr/20Au; $J_1=-1.75\ \mathrm{erg/cm^2}$, $J_2=0.36\ \mathrm{erg/cm^2}$:
(2b) whisker/11Cr/15Fe/11Cr/20Au; $J_1=-1.25\ \mathrm{erg/cm^2}$, $J_2=0.35\ \mathrm{erg/cm^2}$. In the latter structure only the Fe thickness was varied by 3 ML and this change has clearly had an effect on the bilinear coupling strength J_1. Note that the biquadratic coupling term is essentially the same for all four specimens. Since the exchange coupling for Cr/Fe/Cr films depends upon the Fe film thickness, it follows that the Fe/Cr interfaces support the formation of electron resonance states in the Fe films. On the other hand, the Fe/Au interfaces tend to suppress electron resonance states. The full thickness dependence of the exchange coupling in the Fe whisker/Cr/Fe/Cr/Au(001) system will be carried out in a separate study.

TABLE I. The dependence of the bilinear J_1 and biquadratic J_2 exchange coupling parameters on iron film thickness for an Fe-whisker/11 Cr/d Fe/20Au specimen, where d is the iron film thickness in monolayers. J_1 and J_2 are in erg/cm^2; see Eq. (1) of the text.

	BLS		MOKE	
d	$-J_1$	J_2	$-J_1$	J_2
10	0.74	0.23	1.0	0.25
20	1.02	0.23	1.1	0.33
30	0.96	0.23	1.15	0.31
40	0.95	0.23	1.08	0.34

CONCLUSIONS

Fe whiskers provide the best available templates for the growth of Fe/Cr/Fe(001) structures because their surfaces are atomically smooth over regions whose dimensions exceed a micron. By monitoring the RHEED intensity oscillations and specular spot line profiles during the growth one is able to prepare atomically smooth Cr(001) layers for which the last deposited Cr atomic layer grows with a smoothness similar to that of the initial Fe whisker template. Samples grown at the optimum temperature (~300 °C) showed unattenuated RHEED intensity oscillations having well defined cusps at the RHEED intensity maxima. The first monolayer of Cr exhibits a unique behavior; the first RHEED intensity oscillation shows a strong peak having a very sharp cusp even at substrate temperatures as low as 150 °C. This indicates that the first atomic layer is very smooth and reproduces the template. The situation changes when Cr is deposited on a Cr template. In that case the growth of Cr proceeds layer by layer only if the substrate temperature is adjusted to an optimum growth temperature, $T_{s,\mathrm{opt}}\sim 250-300\ ^\circ\mathrm{C}$. In order to obtain a final Cr atomic layer having a low density of atomic steps one must deposit the first Cr layer at a temperature greater than 100 °C. ARAES studies have shown that interface alloying is present at the Fe-whisker/Cr interface even at substrate temperatures as low as 100 °C. This means that the first deposited layer of Cr is atomically smooth but is chemically inhomogeneous due to interface alloying with the Fe substrate as the result of an atom exchange mechanism. It should be noted that this atom exchange mechanism does not operate when Fe is deposited on Cr, so that, in principle, the Cr/Fe interface can exhibit an abrupt change in composition in contrast with the Fe/Cr interface. We showed that the initial stages of the Cr growth strongly affect the strength and the type of exchange coupling. Lowering the substrate temperature during the growth of the first Cr atomic layer helps to increase the strength of the exchange coupling.

BLS and MOKE measurements have shown that the exchange coupling in Fe/Cr/Fe(001) structures having a low density of atomic steps is well described by the sum of bilinear and biquadratic angular terms; Eq. (1). The bilinear exchange coupling strength exhibits short- and long-wavelength oscillations. The long-wavelength oscillations are dominant for Cr thicknesses less than 8 ML. The short-wavelength oscillations are dominant for thicknesses greater than 8 ML. The biquadratic coupling term is always present, but is less than 20% of the bilinear exchange coupling term. The Fe magnetizations are always collinear in the ground state of the Fe/Cr/Fe(001) system: they are either parallel (FM coupling) or antiparallel (AF coupling). It has been pointed out that deviations between measured magnetization loops and magnetization loops calculated using bilinear and biquadratic interfacial coupling terms, Eq. (1), are caused by lateral variations in the exchange coupling parameters. These lateral variations amount to ~15% of the mean value and are characterized by a lateral scale that is smaller than 10 μm. They are very likely caused by interfacial alloying at the Fe/Cr interface. Interface alloying affects several features of the short-wavelength oscillations: (a) The first crossover to antiferromagnetic coupling occurs at 4 ML; (b) the strength of the short wavelength oscillations is weak for Cr thicknesses between 5–9 ML; (c) the phase of the short-wavelength oscillations is reversed from that expected for perfect interfaces and a Cr spacer containing a spin-density wave; (d) the strength of the measured exchange coupling is significantly smaller than that obtained from first-principles electron band calculations. Recent theoretical calculations have been able to explain points (a) and (c). Points (b) are (d) have not yet been fully addressed, but one intuitively expects, and the results of calculations indicate,[14] that a first-principles calculation taking into account interfacial alloying at the Fe/Cr interface would lead to significantly decreased values of the exchange coupling compared to those obtained for perfect interfaces. However the full angular dependence of the exchange coupling should be calculated in order to explain the experimental fact that the coupling can be described by bilinear and biquadratic coupling terms and not by the proximity effect term $(\Delta\theta)^2$ that is expected to be valid for ideal interfaces.

The exchange coupling was not found to depend in any measurable way on the Fe film thickness when the iron film was terminated with a gold layer. This means that the exchange coupling in Fe-whisker/Cr/Fe/Au(001) is not affected by quantum well and resonance states in the Fe film. There is, however, evidence that the thickness of the iron film does affect the coupling strength when the iron surface is bounded by Cr at both surfaces. Experiments on specimens containing a Cr spacer layer and a thin Fe layer capped by Cr will be the subject of future investigations. First-principles calculations are needed to understand why electron standing-wave effects in an iron film terminated by gold appear to be absent compared with those in a thin film terminated by Cr at each surface.

The role played by the interface in exchange coupling between iron films separated by a chromium interlayer was further studied by fabricating specimens having the structure Fe(001) whisker/NCr/nX/20Fe(001)/20Au where N is the number of atomic layers of Cr and n is the number of atomic layers of a different metal X. We used $X=$Cu, Ag, and Mn. The number of Cr atomic layers was restricted to $N=11$ and 12 for these experiments. The results of studies using MOKE and BLS to investigate specimens containing Cu or Ag interface layers indicated that the exchange coupling through atomically smooth Cr layers greater than 8 but less than 24 ML thick can be understood as a combination of a spin-density wave contribution describable by a Heisenberg type of Hamiltonian[14] plus a quantum well contribution due to multiple scattering at the interfaces. This conclusion is supported by the recent first-principles calculations reported by Mirbt and Johansson.[32] However, alloying at the interface between the iron-whisker substrate and the Cr, mediated by an atom exchange mechanism, severely affects the exchange coupling through Cr layers less than 8 ML thick. In this regime the exchange coupling exhibits a large antiferromagnetic background plus a small superimposed short-wavelength contribution. The Fe/Cr transitional region reverses the phase of the short-wavelength oscillation and severely reduces the strength of the coupling relative to that expected for a perfectly sharp interface.

The results of MOKE and BLS experiments using specimens containing Mn interface layers ($X=$Mn) can be understood assuming that the (001) planes of the Mn are fully magnetically compensated. This assumption agrees with recent theoretical calculations[39] for bulk bct Mn having the Fe in-plane lattice spacing. It is also in agreement with magnetic x-ray dichroism and electron capture measurements carried out on Mn grown on an Fe(001) template.[41–43] The phase of the exchange coupling through the Fe-whisker/Cr/Mn/Fe(001) samples was unaffected by the presence of the Mn layers: the strength of the coupling was increased by the factor 2.5 for the case of 1 ML of Mn. The specimen containing three atomic layers of Mn exhibited a noncollinear magnetic ground state and a large contribution to the biquadratic exchange coupling term [Eq. (1)]. This noncollinear ground state and relatively large biquadratic exchange is most likely caused by interface roughness. Uncompensated magnetic moments of the Mn atoms in partially filled atomic terraces can result in magnetic frustration of the Fe magnetic moments at the Mn/Fe interface and this frustration would likely result in a strong contribution to the biquadratic exchange coupling. Further studies of the net magnetic moment in Mn layers are needed, perhaps using magnetic x-ray dichroism, to find out whether the noncollinear ground state in these samples is caused by interface roughness or whether it constitutes an intrinsic property of Fe/Cr/Mn/Fe(001) specimens containing thicker Mn layers.

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