Exchange coupling through spin density wave Cr(001) using Fe whisker substrates

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Exchange coupling through spin density wave in Fe whisker/Cr/Fe(001) structures was studied by Brillouin light scattering (BLS) and longitudinal magneto-optical Kerr effect (MOKE) techniques. It will be shown that interface alloying at the Fe whisker/Cr interface profoundly affects the behavior of short wavelength oscillations. The first crossover to antiferromagnetic coupling occurs at 5 monolayers (ML), the phase of short-wavelength oscillations is reversed compared to that expected for the spin density wave in Cr(001), and the strength of coupling is significantly decreased from that obtained from first principle calculations. Using Cu and Ag atomic layers between the Cr(001) and Fe(001) films, heterogeneous interfaces showed that the exchange coupling in Cr(001) is strongly affected by electron multiple scattering. It appears that electron quantum well states in the Fe film play no important role in the strength of the exchange coupling when the Fe film is bounded on one side by Au, but they become important when the Fe film is bounded by Cr on both sides. © *2000 American Institute of Physics.* [S0021-8979(00)31108-2]

The scanning electron microscopy with polarization analyses (SEMPA) studies by the NIST group[1] using Fe whisker/Cr/Fe(001) samples showed that the exchange coupling oscillates with short wavelength period ~2 monolayers (ML). The SEMPA images revealed in a very explicit way that short wave and long wavelength oscillations exist in the thickness range of 5–80 ML of Cr spacer. The SFU group has carried out quantitative studies using Fe whisker/Cr/Fe(001) samples.[2–4] 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 magnetic optical Kerr effect (MOKE) and Brillouin light scattering (BLS) measurements exhibit two critical fields H_1 and H_2. 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 H and the sample was fully saturated, no asymptotic approach to saturation was observed, see Fig. 1(a). For $H_1<H<H_2$ the magnetic moments were noncollinear, the magnetic moments deviated from the external field direction. For $H<H_1$ the magnetic momentor were antiparallel. In all our samples the field dependence of magnetization loops is consistent with the assumption that the angular variation of the exchange coupling can be expressed in terms of bilinear J_1 and biquadratic J_2 exchange coupling, $E_{\mathrm{exch}}=-J_1\cos(\theta)+J_2\cos^2(\theta)$. The strengths of J_1 and J_2 were determined by a comparison of the experimental data with the theory described in Ref. 5.

The observed approach to saturation at the critical field H_2, see Fig. 1(a), and the onset of the antiferromagnetic configuration at the critical field H_1, see Fig. 1(a), are more gradual than those calculated using the bilinear and biquadratic exchange coupling terms. The calculated approach to saturation clearly exhibits a well-defined kink at the field H_2, the experimental measurements usually show a concave approach to saturation. According to micromagnetic 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 features can be explained by an inhomogeneous distribution of the exchange coupling. A 10% variation in the exchange coupling across the measured area would result in hysteresis loops that are very similar to those obtained using MOKE see Fig. 2.

Quantitative BLS studies have clearly exhibited short wavelength oscillations in the exchange coupling, see Fig. 3. 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.

Several features are important to emphasize. First the exchange coupling crosses to the antiferromagnetic (AF) coupling at 4 ML of Cr. Second, for a Cr spacer thickness $d_{\mathrm{Cr}}<8$ ML the strength of the short wavelength oscillations is quite weak, $|J_1|\sim 0.1\ \mathrm{ergs/cm^2}$. The exchange coupling in this range is only AF due to the presence of an AF long-wavelength bias. This AF bias is peaked around 6–7 ML of Cr. It is interesting to note that the strength of the long-wavelength AF bias is very close to that observed in Fe/Cr/Fe(001) epitaxial multilayers prepared by sputtering where the interface roughness annihilated the presence of the short wavelength oscillations.[6] It follows that the AF bias in Fig. 3 is for the first 7–8 ML of Cr is most likely due to long wavelength oscillations in 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 AF coupled, $J_{\mathrm{tot}}=|J_1-2J_2|\sim 1.0$–$1.5\ \mathrm{ergs/cm^2}$, for an odd number of Cr atomic layers and ferromagnetically (FM) coupled for an even number of Cr atomic layers, $J_{\mathrm{tot}}\sim 0.2\ \mathrm{ergs/cm^2}$.

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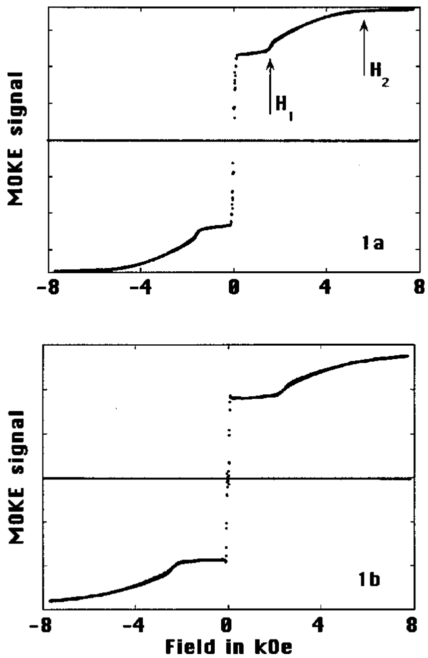

FIG. 1. The magnetization along the applied external field (longitudinal MOKE). The samples were grown using the same Fe whisker substrate using a shutter during Fe deposition. (a) Fe whisker/11Cr/15Fe/11Cr/20Au(001). The integers represent the number of atomic layers. The saturation magnetization was 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 ergs/cc for the bulk Fe and 3.3×10^5 ergs/cc for the thin film. The cubic anisotropy in the Fe film was calculated using formula (1.40) in Ref. 2. The strength of the bilinear and biquadratic exchange couplings $J_1=-1.25$ ergs/cm^2 and $J_2=0.35$ ergs/cm^2, respectively, were calculated using a proper micromagnetic treatment (Ref. 5) for Fe whisker/spacer/FM film systems which includes the rotation of the saturation magnetization inside the bulk Fe whisker (domain wall). (b) Fe whisker/11Cr/18Fe/11Cr/20Au(001). The cubic anisotropy in the 15 ML Fe film is 3.6×10^5 ergs/cc. $J_1=-1.75$ ergs/cm^2 and $J_2=0.36$ ergs/cm^2. The MOKE signal is not saturated due to the lack of sufficiently high external field.

The coupling between the Fe and Cr atoms at the Fe/Cr interface is expected to be strongly antiferromagnetic[7,8] and in consequence the spin density wave in Cr is locked to the orientation of the Fe magnetic moments. Since the period of short wavelength oscillations is close to 2 ML one would expect AF coupling for an even number and FM coupling for an odd number of Cr atomic layers. Surprisingly both the SEMPA[9] and BLS[10] 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 $J_{max}\sim 1.0$ ergs/cm^2, was found to be much less than that obtained from the first principles calculations,[11,12] $J_1\sim 30$ ergs/cm^2. Our studies showed that the strength of the bilinear exchange coupling J_1 is very sensitive to the initial growth conditions. 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 few Cr atomic layers.[10,13] We demonstrated by using angular resolved Auger electron spectroscopy (ARAES) that atomic interface alloying at the Fe whisker/Cr interface due to an interface atom exchange mechanism plays a very significant role and strongly affects the exchange coupling through the Cr spacer.[3,4,13–15]

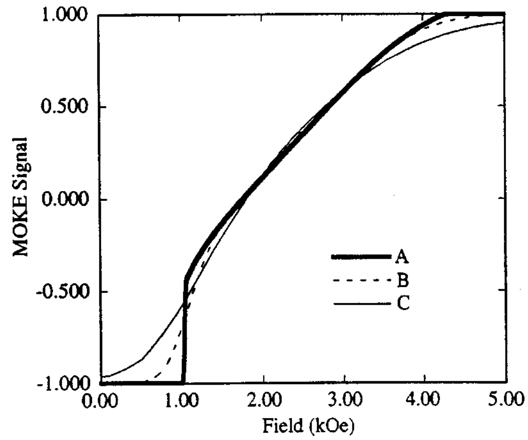

FIG. 2. Calculated MOKE signal for a 20 ML Fe(001) film exchange coupled to a bulk Fe(001) substrate and assuming an inhomogeneous distribution of the bilinear coupling strength J_1. The biquadratic coupling strength, J_2, has been set equal to 0.3 ergs/cm^2 for all curves. The probability of a coupling 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.

The ARAES studies[13,14] showed that the interface alloying during the growth starts already at low substrates temperatures, $T_{sub}\sim 150$ °C involving the top Fe atomic layer. The interface alloying increases with an increasing substrate temperature and at $T_{sub}=300$ °C in the top and first subsurface Fe atomic layers the Fe atoms are partially replaced by Cr atoms with the concentration of 30% and 20%, respectively. It should be noted that interface alloying is driven by the difference in binding energies and is not, in general, symmetric, it occurs chiefly at one interface.

Recently Freyss, Stoeffler, and Dreysse[16] investigated the phase of exchange coupling for intermixed Fe/Cr interfaces. The calculations were carried out by *d*-band tight-binding Hamiltonian using 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 well the experimental studies which were carried out by growing the first few atomic Cr layers at lower substrate

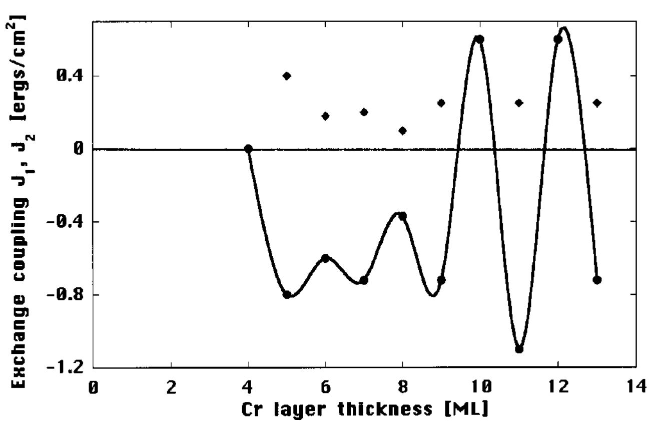

FIG. 3. The thickness dependence of the bilinear J_1 (●) and biquadratic J_2 (◆) exchange coupling. The biquadratic coupling can be measured only for AFM coupled samples. The values of J_2 for the FM coupled samples (10, 12 MLs) was assumed to be equal to those for the AFM coupled samples with 9-, 11-, and 13- ML-thick Cr.

temperatures where the surface alloying is mostly confined to the two interface atomic layers. They found that the reversal in the phase of coupling was predicted already for $x = 0.2$. This result is in very good agreement with our studies. The samples with the Fe/Cr interface prepared at $T_{sub} = 150$ °C, showing only a weak interface diffusion ($x \sim 0.2$),[13,15] had their phase in exchange coupling already reversed from that expected for perfect interfaces.

Heterogeneous Cr spacers were prepared for testing the effect of interface composition on the exchange coupling strength. Fe whisker/11 Cr/1–2 Cu/Fe(001) and Fe whisker/11Cr/1–2Ag/Fe specimens were grown, where the integers represent the number of atomic layers.[3,4] The behavior of exchange coupling in Fe whisker/11Cr/1–2Cu/Fe(001) samples is most surprising. The strength of the exchange coupling in these samples was increased twofold compared to that observed in samples having simple Cr interfaces. Mirbt and Johnson presented calculations[17] that were in accord with our results. Their calculations show that the enhanced coupling strength in Fe/Cr/Cu/Fe(001) samples is due to an increase in the asymmetry of the spin-dependent reflectivity of the Cr spacer electrons at the Cr/Cu/Fe interface. The theoretical calculations by Mirbt and Johnson suggested that a proper model for exchange coupling through spin density waves in Cr has to include two contributions: (a) spin-dependent potential due to magnetic moments of antiferromagnetic Cr and (b) spin-dependent potential at the Fe/Cr and Cr/Fe interfaces.

It is appropriate at this point to raise the question whether the magnetic state of ultrathin Cr(001) layers grown or Fe whiskers is in an intrinsic spin density wave state. In fact the above MOKE and BLS results are also consistent with the behavior that could be expected from a paramagnetic Cr. The NIST group using SEMPA and a wedge Cr layer grown on Fe whisker template were able to study directly the Cr magnetic moment with increasing Cr thickness.[18] They found that the magnetic moment of surface Cr atoms exhibits short wavelength oscillations corresponding to transversal incommensurate spin density wave. These results were also found to be fully consistent with the behavior of oscillatory exchange coupling in Fe whisker/Cr/Fe(001). It follows that the ultrathin films of Cr grown on Fe whiskers are indeed in the transversal spin density wave state.

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[19,20] such states are not only restricted to nonmagnetic spacers, but are also present inside the ferromagnetic layers, and the coupling cannot be described as the interaction which is solely localized in the interfaces. A strong oscillatory behavior, with a period of 5–6 ML, of the exchange coupling with the Fe film thickness in Fe/Cr/Fe multilayers was found by Okuno and Inomata.[21] In their studies Fe/Cr/Fe(001) samples had rough interfaces and exhibited only long wavelength oscillations with the Cr thickness. Since our interface studies showed that a significant part of the exchange coupling depends on the multiple scattering inside the Cr spacer we measured the thickness dependence of the exchange coupling in Fe whisker/Cr/Fe(001) samples where the thickness of the Fe layer was systematically varied by using Fe wedge samples. We found no evidence for an oscillatory behavior of the exchange coupling as a function of the Fe layer thickness for samples covered directly by Au(001). However, our recent experiments indicate that the exchange coupling is sensitive to the film thickness when the Fe film is first capped with Cr producing the structure Fe whisker/Cr/Fe/Cr/Au(001). We prepared two samples on the same Fe whisker substrates: (a) Fe whisker/11Cr/15Fe/11Cr/20Au(001) and (b) Fe whisker/11Cr/18Fe/11Cr/20Au(001). For sample (a) the exchange coupling strength was found $J_1 = -1.25$ ergs/cm^2, $J_2 = 0.35$ ergs/cm^2, see Fig. 1(a), and for sample (b) $J_1 = -1.75$ ergs/cm^2, $J_2 = 0.36$ ergs/cm^2, see Fig. 1(b). Clearly changing the Fe layer thickness just by 3 ML resulted in a substantial change in the strength of the exchange coupling. We also prepared a sample with an Fe wedge film. The Fe film thickness was in the range between 15 and 23 ML. The preliminary results of BLS data show that the strength of the exchange coupling has a long wavelength oscillatory period of 5–6 ML with maximum of the coupling around 16–17 and 22–23 ML, respectively. Since the exchange coupling in Fe/Cr/Fe/Cr/Au(001) depends on the Fe film thickness, it follows that the Fe/Cr interfaces support the formation of electron resonance states in Fe films. On the other hand Fe/Au(001) interface tends to suppress electron resonance states.

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