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Role of interface alloying in Fe whisker/Cr/Fe(001) structures, angular-resolved Auger electron and MOKE studies

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Abstract

Angular-resolved Auger electron spectroscopy (ARAES) studies were carried out at Fe whisker/Cr(001) interfaces. The interfaces were prepared at 100, 180, 246 and 296°C. Interface alloying increases progressively with increasing substrate temperature. The MOKE measurements on Fe whisker/Cr/Fe(001) showed a strong dependence of the bilinear exchange coupling on interface alloying.

1. Introduction

Fe whisker/Cr/Fe(001) systems have played a crucial role in the study of exchange coupling between two ferromagnets separated by a non-ferromagnetic spacer. The SEMPA studies by the NIST group [1] and the MOKE measurements by the Philips group [2] using Fe whisker/Cr/Fe(001) samples showed that the exchange coupling oscillates with short wavelength oscillations of ~2 ML; see also [3]. The SFU group has 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 this strength with ab initio calculations which included explicitly the presence of spin-density waves in the Cr [6,7]. The requirement of smooth interfaces limited the 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 in Fe whisker/Cr/Fe(001) samples was found to be reproducible only in those structures that exhibited 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 results. The width of the RHEED specular spot profiles had to be monitored and one had to establish conditions such that the atom island formation followed a prescribed pattern of nucleation and growth. The best results were obtained for the case when the spot profiles oscillated repeatedly between narrow peaks (filled atomic layers) and split intensity peaks (half-filled layers). This was possible to achieve by maintaining the substrate temperature in a narrow range of temperatures, $280°C < T_{s,opt} < 320°C$.

The first monolayer of Cr exhibits a unique behavior; the first RHEED intensity oscillation shows a strong peak with a very sharp cusp 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, $T_{s,opt}$.

Quantitative Brillouin light scattering (BLS) studies on Fe whisker/Cr/Fe(001) samples have clearly exhibited short wavelength oscillations in the exchange coupling [4,5]. These studies showed also that the exchange coupling through Cr(001) contains both an oscillatory bilinear, J_1, and positive biquadratic, J_2, exchange coupling terms. The exchange energy is given by

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

where θ is the angle between the magnetic moments of the ferromagnetic layers.

The coupling between the Fe and Cr atoms at the Fe/Cr interface is expected to be strongly antiferromagnetic [8,9] 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 expects AF coupling for an even number of Cr atomic layers and FM coupling for an odd number of

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Cr atomic layers for low coverages of Cr. Surprisingly the SEMPA [10] and BLS [11] measurements showed that the phase of the short-wavelength oscillations is exactly opposite to that expected. This means that the first phase slip occurs already at very low coverages of Cr (> 3 MLs). It is also important to note that the strength of the exchange coupling $|J_{max}| \approx 1.0$ erg/cm^2, was found to be much less than that obtained from first-principles calculations, $|J_1| =$ 30 erg/cm^2 [6]. This represents a significant disagreement between experiment and theory. Since Fe whisker/Cr/Fe(001) samples can be prepared in a nearly perfect layer-by-layer growth, the above disagreement between the measured exchange coupling and the theoretical calculations can not be a priori blamed on poor sample quality.

Our recent 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 Cr atomic layer. This behavior led us to believe that the atomic formation of the Cr layer is more complex than has so far been acknowledged. In the following we will demonstrate by using angular-resolved Auger electron spectroscopy (ARAES) that atomic interface alloying at the Fe whisker/Cr interface plays a very significant role and strongly affects the exchange coupling through the Cr spacer.

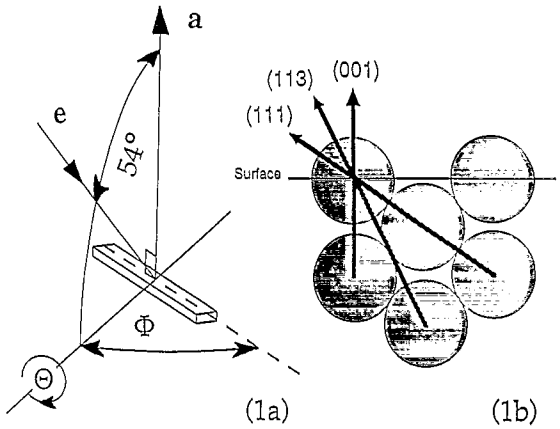

Fig. 1. Experimental configuration for angular-resolved Auger electron spectroscopy (ARAES) measurements. a is the line of acceptance of Auger electrons, and the incident electron beam (2 keV) is directed along the line e. The angle between a and e is 54°, and n is the sample surface normal. The drawing shows the configuration for $\Theta = 0°$, n parallel to a. Angular-resolved studies were carried out using the polar angle Θ (measured from n). The azimuthal angle Φ is used to select an appropriate lattice plane for the ARAES measurements. An illustration of rows of atoms in the bcc (011) crystallographic plane ($\Phi = 45°$), which dominantly contribute to forward scattering of Auger electrons, see details in the text.

2. ARAES studies

The angular distribution of the Cr LVV Auger electrons (529 eV) was used to investigate interface alloying at the Fe whisker/Cr(001) interface. Auger electrons having an energy of several hundred eV energy are strongly forward scattered in the directions interconnecting the emitter with its nearest- and next-nearest neighbors [12–14]. The experimental configuration in our ARAES studies is shown in Fig. 1. The ARAES studies were carried out using an uncommon whisker blade, $100 \times 500 \times 10^4$ μm, with the [001] axis along its long edge and with the large facets oriented in {001} planes. The Fe whisker was mounted on a UHV goniometer equipped with polar, Θ, and azimuthal, Φ, rotation degrees of freedom, see Fig. 1a. The line of acceptance of Auger electrons, a, to the OMNI lens of hemispherical analyzer, PHI-10-360 (2° angle of acceptance with a focal area $\phi = 500$ μm), and the normal to the sample, n, lies in the plane perpendicular to the polar axis, Θ, of our UHV goniometer. In our ARAES studies the Fe whisker was initially oriented with its (011) crystallographic plane perpendicular to the polar axis corresponding to the azimuthal angle $\Phi = 45°$. The incident electron beam (2 keV), e, lies in the plane formed by the polar axis and a, and makes the angle of 54° to a. The angular dependence of the LVV Cr and Fe Auger peaks was measured by rotating the sample, using the polar angle Θ of our UHV goniometer. In this configuration the Auger electrons originating in the top (1st) surface layer have a flat angular dependence. The atoms in the 2nd, and 3rd atomic layers have Auger peaks enhanced for polar angles $\theta = 54.7°$ (along body diagonal [111]); the Cr atoms in the 3rd and 4th atomic layers are enhanced for $\Theta = 0°$ (along the cube edge [001]); the Cr atoms in the 4th atomic layer are enhanced for 25.2° (along the crystallographic orientation [113]: see Fig. 1b). Thus the Cr occupation of the individual atomic layers near the surface can be determined by measuring the polar angular dependence of the Cr KVV (529 eV) Auger peak. The orientation of the incident electron beam, e, in our system leads to a more complex behavior when the Auger signal is measured along the [001] axis. For $\Theta = 0°$ the impinging electron beam is nearly parallel to the body diagonal. In this case the flux of impinging electrons is significantly enhanced for those atoms which are in the 2nd and 3rd atomic layers. The effect is caused by the forward focusing effect due to the row of atoms along the body diagonal. The maximum around $\Theta = 0°$ could be incorrectly interpreted as evidence for the presence of Cr atoms in the 3rd layer. This spurious effect was eliminated by rotating the azimuth Φ of the sample away from 45° to 34.2°. The 45° azimuth was perfectly suitable for measurements that are away from the sample normal, $\Theta > 10°$, where the forward focusing effect is absent.

The goal of our ARAES measurements was to identify the level of interface Fe–Cr alloying. For this purpose we

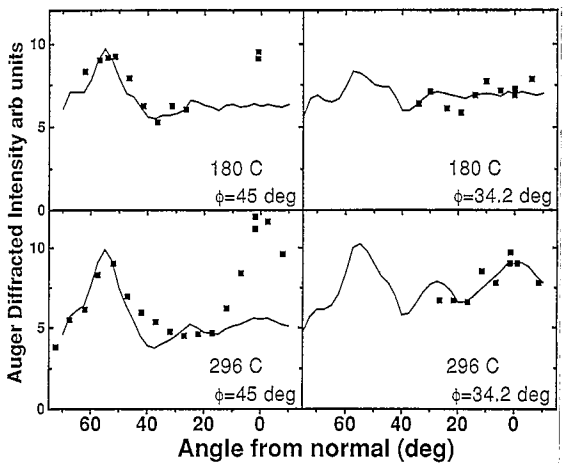

Fig. 2. Polar angular dependence of the LVV Cr Auger electron peak from a 0.5 ML thick Cr layer deposited on an Fe(001) whisker blade. The deposition of Cr was carried out at the substrate temperatures of 180 and 296°C, respectively. The peaks around $\Theta = 0°$ for the azimuth $\Phi = 45°$ are caused by forward focusing effect of the incident electron beam, see the text.

deposited 0.5 ML of Cr at various substrate temperatures and carried out appropriate ARAES using the Cr KVV Auger peak. The results of our studies are shown in Fig. 2, where the ARAES data are shown for two substrate temperatures, $T_s = 180$ and 296°C. More detailed presentation can be found in Ref. [15]. The measurements for $T_s = 180$°C show no maximum around $\Theta = 0°$, but a clear maximum is visible for $\Theta = 54.7°$. The results for $T_s = 296$°C show maxima for both angles. The presence of these maxima show clearly that interface mixing is present during the growth of Cr on Fe whisker substrates. At 180°C the Cr atoms only penetrate into the first atomic layer below the surface of the Fe. At 296°C the Cr atoms even enter the second atomic layer below the surface. No peak is evident along 25° indicating that in this range of substrate temperatures the Cr ad atoms do not penetrate into the 4th atomic layer. The measured ARAES data were analyzed using the computer program 'ssc' developed by Fadley and coworkers [14]. The scattering is treated using scattering phase shifts from the computer program 'FEFF-3' developed by Rehr and coworkers [16]. The single scattering approach is fully justifiable for Cr atoms distributed in the first two atomic layers. For atoms in the third atomic layer the single scattering approach is not fully adequate around $\theta = 54.7°$. In this case the Auger electrons propagate along two atoms in the body diagonal where the effect of multiple scattering is not negligible. Calculations along the close-packed directions show that the effect of multiple scattering is equivalent to the reduction of the intensity of the Auger electrons to 60% of its value calculated using the single scattering approach [14]. The results of our analyses are summarized in Table 1, which shows that interface alloying plays a role even at low substrate temperatures and becomes very significant at substrate temperatures for which a thick Cr spacer can be grown in the layer-by-layer mode. One should point out that interface alloying is an asymmetric effect; it happens only at one interface [17].

Table 1
Fractions of the Cr atoms in the 1st (surface layer), 2nd and 3rd atomic layers obtained by 'ssc' fits [14]

Temperature (°C)	1st	2nd	3rd
100	0.82	0.13 (0.18)	0.05
180	0.74	0.21 (0.26)	0.05
246	0.67	0.30 (0.33)	0.03
296	0.51	0.30	0.19

A half ML of Cr was grown on an Fe(001) whisker template at 100, 180, 246, and 296°C. The use of a 3rd layer in 'ssc' calculations for samples prepared at 100, 180 and 246°C provide better overall fits around $\Theta = 0°$, but the measured angular variations around $\Theta = 0°$ are on the borderline of our experimental accuracy, and therefore the fractions determined in the 4th column might well be incorporated into the 2nd layer (shown in brackets in the 3rd column).

3. Magnetic studies and discussion of results

The exchange coupling through Cr is most likely affected by interface alloying. This point was verified by using two Fe whisker/11Cr/20Fe(001) samples. Thicknesses are in ML. In the first sample the Fe/Cr interface was formed at $T_s = 180$°C and then the rest of the Cr spacer was grown with $T_{s,opt}$ to maintain a good layer-by-layer growth at higher Cr coverages. The second sample was grown by depositing 7 ML at $T_s = 246$°C and then the

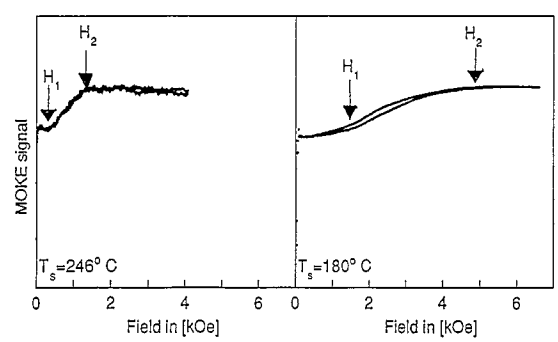

Fig. 3. MOKE signal for Fe whisker/11Cr/20Fe/20Au(001) samples prepared with the first Cr atomic layer grown at $T_s = 180°$ and 246°C, respectively, see details in the text: thicknesses are measured in ML. A lower substrate temperature (180°C) during the growth of the first Cr atomic layer leads to a significantly larger value of J_1 than that observed for the sample prepared at a higher substrate temperature (246°C). The results of computer analyses led to $J_1 = -0.41$ erg/cm^2, $J_2 = 0.17$ erg/cm^2 for $T_s = 246°$; and $J_1 = -1.23$ erg/cm^2, $J_2 = 0.27$ erg/cm^2 for $T_s = 180$°C.

last 4 ML were grown at $T_s = 294$°C. The results of magnetic MOKE studies are shown in Fig. 3. The MOKE measurements show 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, the sample is fully saturated, no gradual approach to saturation is observed. For $H_1 < H_2$ the magnetic moments were non-collinear, the magnetic moments deviated from the external field direction. The anti-collinear configuration is reached below the second critical field H_1, $H < H_1$. The existence of a fully saturated state for fields $H > H_2$ is consistent with the assumption that the angular variation of the exchange coupling is expressed in terms of bilinear and biquadratic exchange coupling, Eq. (1). The separation between H_2 and H_1 fields requires the presence of J_1 and J_2. The positions of the critical fields H_1 and H_2 were calculated using a fully micromagnetic calculation for an Fe whisker/Cr/20Fe sample [18]. The results of these calculations show that the exchange coupling depends very strongly on interface alloying. The sample with the Fe/Cr interface prepared at a lower substrate temperature, $T_s =$ 180°C, exhibits a much larger bilinear exchange coupling, $J_1 = -1.23$ erg/cm^2, than the specimen having an Fe/Cr interface prepared at a higher substrate temperature, $T_s =$ 246°C, $J_1 = -0.41$ erg/cm^2. The biquadratic exchange coupling was found to be only weakly dependent on the substrate temperature during the growth of the first Cr atomic layer; $J_2 = 0.17$ erg/cm^2 for $T_s = 180$°C, and $J_2 = 0.27$ erg/cm^2 for $T_s = 295$°C.

Stoeffler and Gautier have shown that the presence of Fe in Cr atomic layers around the interfaces affects the magnetic behavior [19]. Tight-binding calculations for $[Fe/Cr/Fe]_n$ superlattices, which have their interfaces formed by a two ML ordered alloy {Fe(75%)–Cr(25%); Fe(25%)–Cr(75%)}, showed a marked difference in the orientation of the magnetic moments at the interfaces. The Cr moment in the Cr-rich layer, Fe(25%)–Cr(75%), is oriented parallel to the adjacent pure Fe layer, and the first 'clean' Cr atomic layer is oriented antiparallel to the Fe layer. Effectively this results in the change of the number of Cr atomic layers by one in our whisker/Cr/Fe specimens. An odd number of deposited Cr layers becomes effectively an even number of Cr layers and vice versa. Under these circumstances the phase of the short wavelength oscillations, in samples where only one interface is effected, is reversed from that expected from theoretical calculations using abrupt interfaces. The strength of the coupling in these calculations was found to be decreased substantially. The interface alloying has indeed a very pronounced effect on the magnitude of the bilinear exchange coupling, see Fig. 3, and therefore the observed change in the phase of the short wavelength oscillations suggests very strongly that the magnetic state of the alloyed atomic layers is responsible, in a similar manner to that in Stoeffler and Gautier calculations, for this phase slip. Extensive studies are under way to find out whether the degree of interface mixing can be limited to the point where the measured phase of the short wavelength oscillations can be reversed and brought into agreement with the first principles calculations.

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