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Electronic aspect and giant magnetoresistance in Co/Cu/Co sandwich structures

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Abstract. We have studied the electronic states in a non-magnetic spacer layer of Co/Cu/Co sandwich layers using the magneto-optical Kerr effect (MOKE) and x-ray photoemission and have found that they are discrete owing to the quantum size effect and appear approximately every 10.6 ± 0.5 Å. This coincides with the oscillation period of the saturation field. We connect them with the quantum minority-spin interface states near the Fermi level E_F . The magnetoresistance found in these sandwich layers has been calculated on the basis of the quantum size effect transport theory with the requirement that there exist quantum well states within individual layers or groups of layers. The results of the calculation and experiments have been discussed.

Among metallic multilayers consisting of an alternating thin magnetic layer and Cu layer which show giant magnetoresistance, the Co/Cu system is an example of particular interest because it displays a very large magnetoresistance even at room temperature [1, 2]. A comparatively large magnetoresistance is still retained until its multilayer structure is reduced to the simplest trilayer form where the Cu layer is sandwiched between two magnetic Co layers. Maximum room-temperature magnetoresistance values as large as 10% have been reported in Co(14 Å)/Cu/Co(15 Å) sandwiches grown on thin Fe buffer layers of thickness 50 Å with Cu layers of thickness between 7 and 50 Å, where the difference between the coercivities of the two magnetic layers enhances the antiparallel alignment of magnetizations, leading to a large magnetoresistance [3, 4].

In ultrathin films, on the other hand, the perpendicular wavevector can be quantized, giving rise to resonances in the density of electron states, due to the electronic potential discontinuities experienced by electron states at interfaces. It has been shown that an epitaxial noble metal overlayer on transition metal, e.g. Cu on FCC Co(100), exhibits sp symmetry electronic states which are spin polarized by the confining magnetic interface [5, 6]. Similar electronic states have also been identified for the MBE-grown epitaxial overlayers and thin films containing noble metals using the magneto-optical Kerr effect (MOKE) [7, 8]. These states present some of the characteristic features of one-dimensional quantum well states. The confinement of electrons in such a system of small spatial dimensions also results in a discrete energy-level spectrum for the response function and thus modifies significantly the electronic transport properties. Of the present sandwich structures their sizes in the perpendicular direction are of the same order of magnitude as or smaller than the mean free path of electrons. What features their electronic structures possess and

how their transport properties can be explained therefore become questions that have to be examined. The purpose of this paper is first to investigate the electronic structures in sputtered Co/Cu/Co sandwiches using MOKE and x-ray photoemission spectroscopy (XPS) measurements. We then extended our investigation to their magnetoresistances including a calculation based on the quantum size effect transport theory. It is interesting to see from the current results that the quantum states, found in MBE-grown epitaxial films with a well refined uniform interface and/or surface on an atomic scale are still observable in the perfect sputtered films and sandwich structures, which affect in turn the magnetoresistance.

The Co/Cu/Co sandwich specimens were deposited in an atmosphere of argon gas at a pressure of 5 mTorr with a sputtering rate of about 1 \AA s^{-1} using an RF magnetron sputtering system having a base pressure of less than about 4×10^{-7} Torr. To prepare the Co(14 Å)/Cu(t)/Co(15 Å) ($t = 7\text{--}50 \text{ \AA}$) sandwich structures, the growth procedure was begun by depositing an Fe buffer layer (50 Å) onto a well cleaned glass substrate. Measurements of the room-temperature MOKE were carried out with both polar and longitudinal geometries on families of specimens from the Co layer surface within a wavelength range 400–800 nm using the polarization angle modulation technique described in more detail elsewhere [9]. Photoemission spectra measurements were performed using an x-ray source of monochromated Al K α radiation (Surface Science Instrument, X-probe) to follow the evolution of the valence band states corresponding to increasing Cu thickness sandwiched by Co layers. The magnetoresistances were measured by means of a standard four-probe method with an applied field of 2.5 kOe. The current and field were arranged in the plane of sandwich structure with the field orthogonal to the current direction. The saturation magnetoresistance is defined as the difference between the maximum resistance and the resistance at saturation field, divided by the latter.

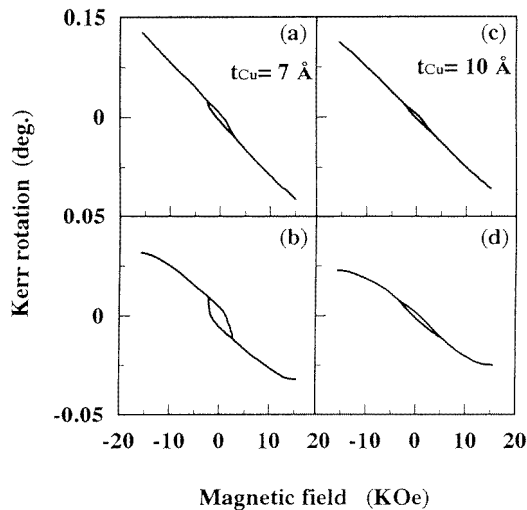


Figure 1. MOKE hysteresis loops measured at a field of 16 kOe, for Fe(50 Å)/Co(14 Å)/Cu(t)/Co(15 Å) sandwich layers with $t = 10 \text{ \AA}$ and 20 \AA , respectively, where loops (b) and (d) represent the parts left of loops (a) and (c) after a withdrawal of the unsaturated contributions from the magnetic layers that change linearly with the field.

To begin with, the room-temperature polar MOKE loops are shown in figure 1 for the Fe-buffered specimens of Co14 Å/Cu/Co15 Å with Cu spacer thicknesses of 7.0 and 10 Å.

They show almost linear variation with magnetic field (figures 1(a) and 1(c)) and remain unsaturated even in a field of 16 kOe, following the fact that the Co and/or Fe layers have in-plane easy-magnetization directions. Figures 1(b) and 1(d) show the MOKE loops for the same specimens after the unsaturated linear components were eliminated. They look like the corresponding loops in figures 1(a) and 1(c) but with the slopes of their loops modified. Note that the contribution to the magnetic moments due to the spin polarization of electrons in the spacer layer, and therefore the magneto-optical activities, are only several per cent of those from adjacent magnetic layers [10]. The information about the electronic structure of the Cu spacer layer should be included in the total MOKE signals obtained from the whole sandwich layers. Unlike the measurements of the longitudinal MOKE, the ability to remove a large part of the contributions from adjacent magnetic layers to the MOKE signals due to their approximately linear variations with magnetic field in this situation favoured greatly the purpose of these experiments because we can study the electronic structure of the Cu spacer layer more clearly. With this procedure for all the data of other specimens measured at different photon energies we reproduce the simplest view in figure 2 of some intrinsic features that might relate to the electronic structure of the systems, and in particular to those of the Cu spacer layer. The features seen in the spectra of both bulk Fe and Co [9] are still pronounced in the spectra of the sandwich structures for both low and high energies (1.6 and 3.2 eV). The dip at about 2 eV in the spectra, which is independent of Cu spacer thickness and does not appear in the spectra of either pure Co or Fe, can be identified as responsible for the magneto-optical activities near the Cu plasma edge. Typically a novel dip is observed at some 2.3 eV in the spectrum of the specimen with a Cu spacer thickness of 7 Å. It shifts to a higher energy with increasing Cu spacer thickness.

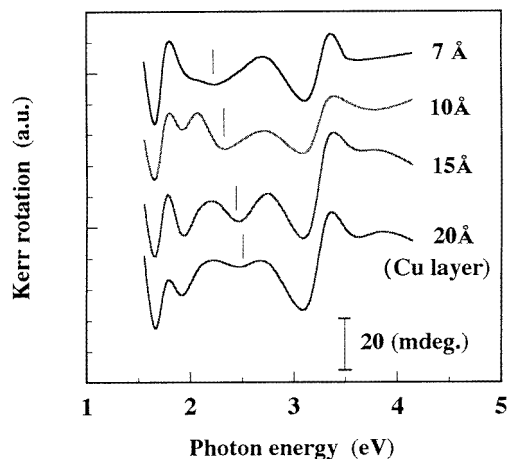


Figure 2. MOKE difference spectra for the same sandwich layers with Cu layer thicknesses equal to 7, 10, 15 and 20 Å, in which the unsaturated contributions from the magnetic layers are removed as shown in figure 1(b) and (d) (a.u., arbitrary units). They are offset for clarity.

To connect this novel magneto-optical transition with the electronic structure of sandwich layers, we examined the absorptive parts of both the diagonal and the off-diagonal conductivities using the data of the saturated Kerr rotation (as seen in figure 2) and Kerr ellipticity, and the virtual optical constants $N = n + ik$ of the sandwich layers. The optical parameters were determined readily by inverting the normal-incidence reflection and transmission function using the effective-reflection approach. By doing this we

clarify the possible vagueness in explaining the direct connection between the MOKE measurements and electronic structure of the sandwich layers because other sources, such as the effects of optical constants and interference in the multilayer structure, might cause an extrinsic enhancement of the MOKE and/or might change the energy dependence of the MOKE spectra [9, 11–12]. The calculated spectra of the absorptive parts of both the diagonal and the off-diagonal conductivities for the specimen with a Cu spacer layer of 10 Å are, for instance, displayed in figure 3. The absorptive part of the diagonal term $(-\omega(n^2 - k^2)/4\pi)$ represents optical dispersion for the whole sandwich structure resulting from both right circularly polarized (RCP) and left circularly polarized (LCP) light in the magnetized metal. The dispersion spectrum resembles that of the Co layer but has the opposite sign to the previous data [13], exhibiting no enhanced dispersion structure given by the Drude model (dispersion due to intraband transitions of conduction electrons) [14]; this indicates interband rather than intraband transitions. Interestingly the initial negative sign of the current data is indicative of the transparent feature of the sandwich layers where the refractive index n becomes larger than the extinction coefficient k , resulting in a high transmittance which generally cannot be expected for bulk metal. This remark, however, is reconciled with the experimental observation on the ultra-thin sandwich layers.

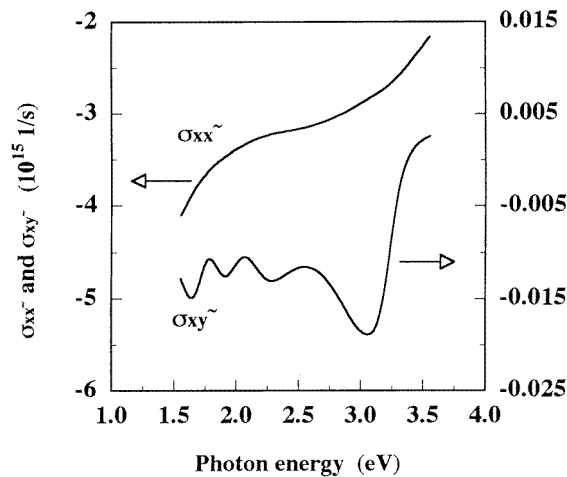


Figure 3. Energy spectra of the absorptive part of the conductivities, σ_{xx}^{\sim} and σ_{xy}^{\sim} (difference spectrum), for Fe(50 Å)/Co(14 Å)/Cu(10 Å)/Co(15 Å) sandwich layers.

While the off-diagonal term, whose absorption spectrum is shown in figure 3, represents the origin of the MOKE in which the difference between the LCP and RCP absorptions plays the leading role but the effects associated with the optical (sum of LCP and RCP) absorption are of secondary importance. It reflects the contribution of the Cu spacer layer and clearly illustrates the enhanced structure characteristic of nearly the same MOKE absorption spectra as shown in figure 2. Concentrating on this spectrum, a well defined novel dip is observed again centred around 2.3 eV amongst the other absorption dips responsible for the plasma edge effect and interband transitions occurring in Co and/or Fe layers. This indicates unambiguously that it is connected with the intrinsic magneto-optical transitions from the 3d band to the sp band near the Fermi level in the Cu spacer layer whose electronic structure is modified by adjacent magnetic layers embedded in the sandwich structures.

The electronic structure of a sandwich structure and the physical properties derived therefrom also depend on its growth orientation. For a (100)-oriented Cu spacer layer, the existence of quantum well states is structurally possible with their wavevectors extending from various edges of the sp band to the Fermi surface since the Fermi level lies in the sp band along the (100) direction (Γ -X line). For the (111)-oriented Cu spacer layer, which prevails mainly in sputtered multilayers with a moderate thickness of several hundred ångströms, there are no sp quantum well states at the Fermi level for the perpendicular wavevector exactly along the (111) direction (Γ -L line) since the Fermi level lies in the sp band gap in this direction. Considering the finite parallel wavevector, one does encounter quantum well states at E_F , but their periods thus determined vary over a wide range. The situation here is more complicated than the above two extremes. The specimens that we used were examined by both a large-angle x-ray scan and TEM diffraction but no particular growth orientations were rigorously found. This was probably due to the limitations of the x-ray diffraction method in not gaining sufficient diffracted energy to detect the structure of these ultra-thin sandwich layers. Nevertheless, our TEM diffraction showed that these sandwich layers have a FCC crystalline structure. Looking more closely at the data on the magneto-optical activities, it seems evident that their behaviours are nearly those of (100)-oriented crystals because there are no such band structures in Cu near the Fermi level except those with Γ -X symmetry that can allow magneto-optical transitions at such a low energy. Similarly, the significant role of the near-(100)-oriented crystalline constituent, even in the (111)-oriented sputtered Co/Cu/Co multilayers, has been shown in determining the antiferromagnetic (AF) coupling which is closely related to the topological structure and symmetry of the Fermi surface [15]. The bulk band structure with Γ -X symmetry therefore is most likely to be suitable for a simple qualitative explanation of our experiments.

As shown schematically in figure 4 [16, 17], the energy gap formed in the vicinity of the Fermi energy level in the Δ_1 band structure of the minority spin of Co(100) due to the s-d hybridization allows the establishment of one-dimensional quantum well states in the minority Δ_1 sp band of Cu(100). In contrast, the majority states in Cu(100) couple with the counterpart component states of the same symmetry in Co(100) and remain a continuum of Bloch states. The tentative assignment of the transition between the minority bands $\Delta_1\{s, p_z\}$ and $\Delta_5\{p_x \pm ip_y, d_{xz} \pm id_{yz}\}$ in the quantum well in Cu(100) gives a transition energy of about 2.3 eV, in fairly good agreement with the observations in the MOKE experiment. Changing the Cu spacer thickness, which is equivalent to changing the thickness of quantum well, further raises the quantum well level particularly relative to the optical transition and consequently leads to a shift in the dip in the spectra of the MOKE absorption to a higher energy.

Displayed in figure 5 is the spacer layer thickness dependence of the MOKE absorption. The Cu-induced structure of the MOKE spectra measured at 2.3 eV is characteristic of oscillation with a period of about 10.6 ± 0.5 Å. As is known, the absorptive part of off-diagonal conductivity and therefore the MOKE absorption are correlated with the density of unoccupied final states and the spin polarization of these initial and final states. In this experiment, we vary the Cu layer thickness, causing the minority-spin state density in it to appear and disappear periodically, while the majority-spin states remain nearly unaffected. Therefore, the oscillation behaviour with Cu spacer thickness of the MOKE absorption is obtained, exhibiting a $\lambda_{env}/2$ ($= 5.8$ layers (10.6 Å)) period as a result of effect of the quantum well states in the Cu spacer layer where λ_{env} represents the wavelength of the envelope wave propagating in the quantum well [5]. When the photon energy is changed, a variation in the oscillation period of the MOKE absorption is observed, e.g. at 3.1 eV. This reflects the fact that the quantum well states cause a oscillation in the MOKE absorption

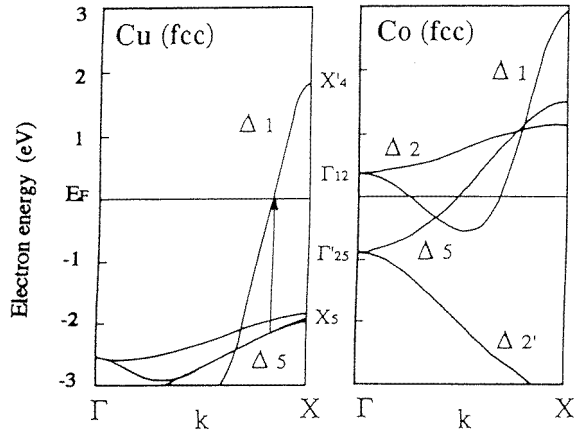


Figure 4. A schematic band structure of bulk FCC Cu(100) (after [7]) and FCC Co(100) (minority-spin electronic band, after [8]). The minority-spin state in Cu exhibits quantum well behaviour at the Fermi level, while the majority-spin Δ_1 states couple with the counterpart states in Co.

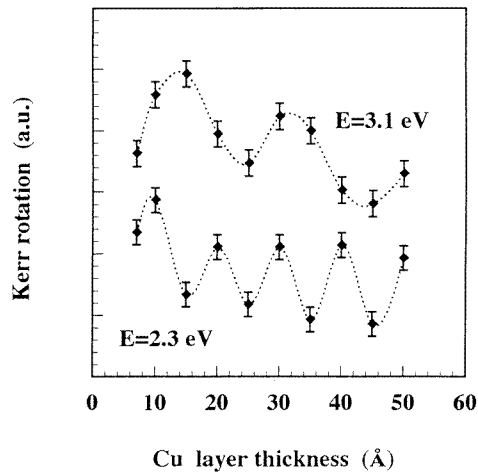


Figure 5. Cu layer thickness dependence of the MOKE absorption at photon energies of 2.3 and 3.1 eV (a.u., arbitrary units). The former corresponds to the optical transition from Δ_5 to Δ_1 minority-spin states at the Fermi level.

with its period modified by the perpendicular wavevector which is in fact photon energy dependent [18].

Spin-integrated photoemission spectra of the valence band for the sandwich structures are presented in figure 6, measured with the 1.48 KeV Al $K\alpha$ x-ray source. The spectra show the Co and/or Fe 3d emission near the Fermi level with an intense structure featured by a Cu 3d-derived peak at 3 eV binding energy whose emission intensity increases with increasing Cu layer thickness. Plotting photoemission intensity, normalized by the Cu 3d-derived peak intensity, at the Fermi level E_F as a function of Cu layer thickness, one obtains the data points in figure 7. The data involve the contribution from Cu sp emission and from

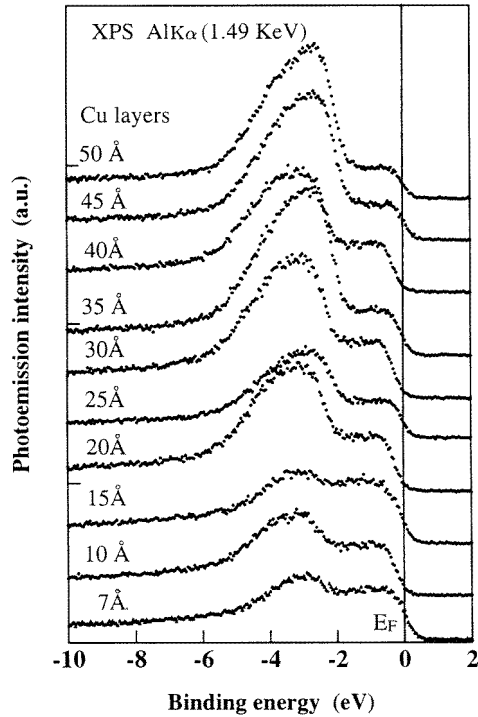


Figure 6. Spin-integrated photoemission spectra of the valence band for sandwich layers with Cu layer thicknesses ranging from 7 to 50 Å (a.u., arbitrary units). The sp band continuum of bulk Cu is discretized into a quantum well state in the Cu spacer layer.

Co 3d emission, exhibiting a modulation of the normalized intensity with a periodicity of 10.6 ± 0.5 Å. Because the delocalized Cu sp-derived states are sensitive to the size of Cu layer thickness, the observed structures can be explained by taking into account the discretization of energy and perpendicular wavevector due to the finite size of the Cu spacer layer. With increasing Cu layer thickness, the electronic states of the sp minority electrons of Cu, partially hybridized with the Cu 3d states, change periodically at the Fermi level E_F , and result in an oscillating photoemission intensity [6]. The resulting modulation periodicity is about 10.6 ± 0.5 Å equivalent to that of the MOKE absorption due to the transition from the Cu 3d states to the sp quantum well states at the Fermi level, corresponding to a photon energy of 2.3 eV (figure 3). The MOKE spectra are in fact related not only to optical and photoemission results but also to the electron spin-polarization effect. Hence this suggests the same physical origin for the oscillating feature found in these two results, i.e. the development of the polarized quantum states of minority electrons in the Cu spacer layer.

To extend the analysis of in-plane electron transport observed in the sandwich layers, we plot in figure 8 the curves of magnetoresistance versus field for Fe(50 Å)/Co(14 Å)/Cu(t)/Co(15 Å) specimens with $t = 10$ and 20 Å, respectively. The variation in the in-plane magnetization behaviours of the two different magnetic layers with the field, probed via the longitudinal MOKE method previously described [4], are presented in figure 9. MOKE measurements of this kind enable us to make specific assignments for the origin of magnetoresistance. Comparison of figures 8 and 9 shows that for the thin

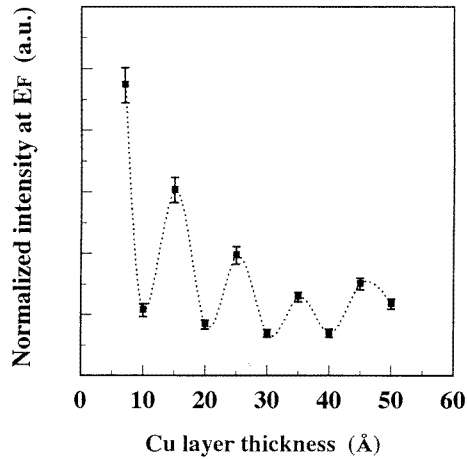


Figure 7. Normalized intensity at the Fermi level versus Cu layer thickness as shown in figure 6 (a.u., arbitrary units). Periodic minima correspond to quantum well states crossing the Fermi level.

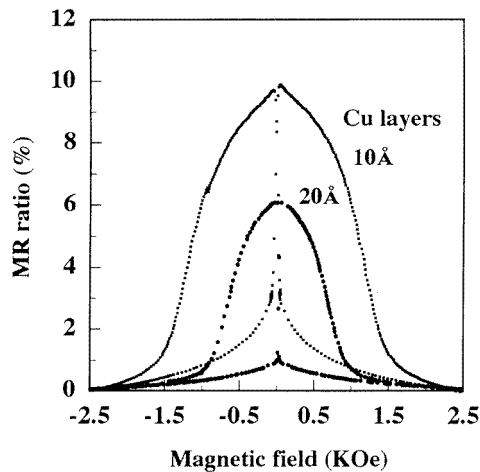


Figure 8. Magnetoresistance (MR) versus field for Fe(50 Å)/Co(14 Å)/Cu(*t*)/Co(15 Å) sandwich layers with Cu spacer layer thicknesses being 10 and 20 Å.

Cu layer thickness (10 Å) both the presence of (AF) coupling and the difference between the coercivities of the two magnetic layers act to enhance the magnetoresistance up to its maximum of 10%. For spacer layers of average thickness (greater than 10 Å) the difference between the coercivities of the two magnetic layers is of prevailing importance in creating the relatively large magnetoresistance since the amount of AF coupling is not strong enough to maintain the antiparallel alignment of magnetizations in the two magnetic layers. The data on the magnetoresistance versus Cu spacer layer thickness is summarized in figure 10(a). If the Cu spacer layer thickness is less than 10 Å, one of the two magnetic layers may couple ferromagnetically with the other through the pinhole in the Cu spacer layer as shown in figure 9. This makes the antiparallel alignment for the two magnetizations collapse, thereby

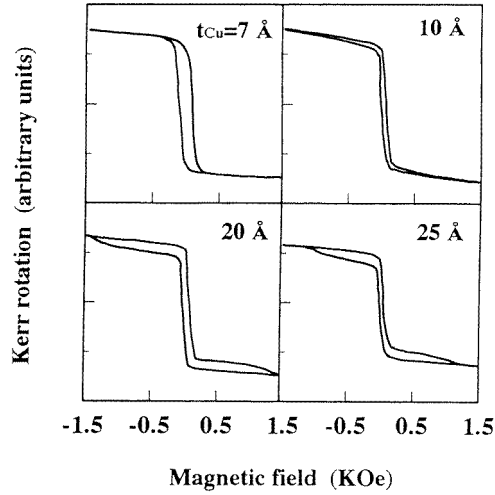


Figure 9. Longitudinal MOKE hysteresis loops measured at a field of 1.5 kOe for for Fe(50 Å)/Co(14 Å)/Cu(t)/Co(15 Å) sandwich layers with $t = 7, 10, 20$ and 25 Å.

leading to an abrupt drop in magnetoresistance.

In the following we treat the data on magnetoresistance in the sandwich layers within the picture of quantum well states. Calculation of magnetoresistance using the quantum well states model [19] assumed that antiparallel alignment of magnetizations is completely realized and the change in magnetoresistance with Cu spacer layer thickness as a whole is attributed to the quantum size effect in the present structures. Further the vacuum-metal-like feature of the interface between the magnetic (M) and non-magnetic spacer layer (N) is retained and the quantum well states exist within each layer and/or groups of layers in these sandwich structures. We take majority- and minority-spin electrons to be separate and distinct spin bands for the purpose of simplifying the calculation of electrical conductivity which is estimated in the presence of both impurity and roughness scattering for a thin film with its z -directional energy-level spectrum quantized. In the ferromagnetic configuration of two magnetic layers, as mentioned above, the majority N electrons couple with their counterparts in the magnetic layer so that they all are indistinguishable from one another and thereafter perfectly transmissive at the N–M interface, while the minority N electrons are distinguishable from minority M electrons, experience confinement within the Cu spacer layer thickness and are perfectly reflecting at the same interface. The conductivity corresponding to this configuration is then obtained as a summation of those of two distinct parallel films $t_{M1} + t_{M2} + t_N$ and t_N thick where t_M and t_N represent the thicknesses of the magnetic and non-magnetic layers, respectively. In the same fashion, we can see that for the AF configuration of two magnetic layers the conductivity can be written as the contribution from two distinct parallel films $t_{M1} + t_N$ and $t_{M2} + t_N$ thick. The magnetoresistance is therefore given by

$$\frac{\Delta R}{R} = \frac{\sigma_F}{\sigma_{AF}} - 1$$

in which we used the value of 1.36 \AA for the Fermi wavevector k_F of Cu and left the impurity mean free path l_0 and surface or interface roughness, i.e. a root mean square deviation around the average film thickness δd , as fitting parameters.

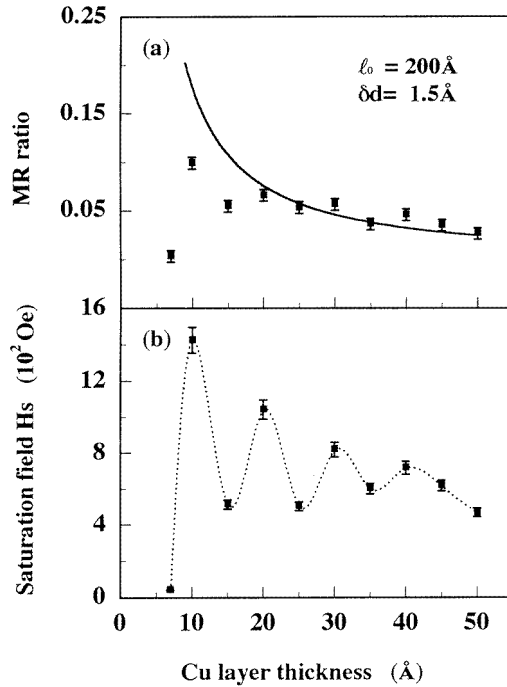


Figure 10. (a) Magnetoresistance (MR) versus Cu layer thickness for the sandwich layers of Fe(50 Å)/Co(14 Å)/Cu(*t*)/Co(15 Å). Calculated results are represented by the solid curve with the fitting parameters of mean free path $l_0 = 200 \text{ \AA}$ and interface roughness $\delta d = 1.5 \text{ \AA}$, and the measured data by the full squares. (b) Saturation field versus Cu layer thickness. The dotted curve gives a guide to the eye. It shows a period of $10.6 \pm 0.5 \text{ \AA}$ for magnetic oscillations, which agrees with the periods found in the MOKE and spin-integrated photoemission measurements.

The calculated result is represented by the solid curve in figure 10(a), where the parameters l_0 and δd are given as 200 \AA and 1.5 \AA , respectively. The value of the mean free path is estimated to be nearly half the value of the mean free path in Cu [20]; the value of interface roughness locates in the range of the data obtained in spin-valve layered structures [21]. In this calculation it was recognized that the amplitude of the magnetoresistance and its spacer layer dependence are strongly affected by the surface or interface roughness rather than impurity scattering, implying a predominant role of spin-dependent interfacial scattering of electrons for the mechanism of magnetoresistance effect in the present sandwich layers. In addition, the calculated resistivities of these sandwich structures fall within the scatter of their corresponding experimental data. They are for instance 30.2 and $21.5 \mu\Omega \text{ cm}$ compared with the measured values of 38.5 and $24.6 \mu\Omega \text{ cm}$, respectively, for Cu spacer layer thicknesses of 7 \AA and 30 \AA . These results are in fairly good agreement with the experiment in estimating the magnitude of magnetoresistance and the trend of its variation with the spacer layer thickness, but not at the expense of physical reasoning of the size of the fitting parameters.

It should also be noted that there seems to be a small oscillating variation in the magnetoresistance with Cu spacer layer thickness, and in phase with the oscillations of the properties mentioned above. The variation, however, is within the experimental error so that we should not prematurely infer definitely that it reflects the intrinsic physical features,

although it is possible in principle. Rather some more accurate measurements on electron transport are needed to elucidate this issue. We believe that electron transport is essentially sensitive to the interfacial conditions, and to some extent the interfacial roughness will blur the fine features of resistivity and magnetoresistance due to the quantum size effect. Moreover, in our simplified calculation, only an envelope function of magnetoresistance versus the Cu spacer layer thickness was obtainable, where the possible oscillating features of electron transport were not taken into account since we had assumed a completely antiparallel alignment of the two magnetizations throughout this treatment. In fact, as a function of the angle between the two magnetizations and therefore the thickness of magnetic layer thickness, the quantum nature of the states in the sandwich layers produces a very sharp peak in the variation in resistivity [22] and a quasi-oscillatory magnetoresistance [23]. Further improvements on the foregoing assumptions then became desirable so as to reflect elegantly the detail of the transport experiments. Even so, the results of these two model calculations, based on the idea of quantum size effects, are not applicable directly to the current data because the high degree of modelling in the calculations makes them far from the experimental limitations. Other approaches to generalize the problem more fully are still required.

A plot of the saturation field measured from the curves of magnetoresistance versus field is displayed as a function of Cu layer thickness in figure 10(b). Again there is a well defined oscillation with a period of about $10.6 \pm 0.5 \text{ \AA}$. On comparison of the magnetic oscillation with the oscillation in the density of states at the Fermi level found by photoemission, both were found to exhibit the same periodicity. The result agrees fairly well not only with the other experimental data [24] but also with those expected by quantum theory [5], which indicates a connection between the magnetic coupling and quantum well states. The period is explained by the fact that the Fermi level crossing of the sp band is close to the band edge at X, which is the origin of k_{env} in the quantum well picture. This is identical with the results of RKKY theory, which predicts that the oscillation period is given by extremal Fermi surface spanning vectors perpendicular to the surface [25]. The results for the (100)-oriented Cu spacer layer in RKKY theory corresponds to our experimental data where the short period of 4.6 \AA predicted by theory did not occur because the interfacial roughness in the sputtered sandwich layers might average it out [26].

In summary, we have studied the electronic states in the non-magnetic spacer layer of Co/Cu/Co sandwich structures and showed that the presence of quantum well states plays a predominant role in determining their magnetic properties and electronic transport. They appear every $10.6 \pm 0.5 \text{ \AA}$, which coincides with the oscillation period of the saturation field in the sandwich structures. Magnetoresistances in these sandwich structures are demonstrated and can be explained fairly well on the basis of spin-dependent electron scattering by impurities and/or at interfaces within a picture of quantum well states.

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