The role of molecular beam epitaxy in research on giant magnetoresistance and interlayer exchange coupling

by R. F. C. Farrow

In this paper, we review the contributions which MBE (molecular beam epitaxy) has made to the field of GMR (giant magnetoresistance) and interlayer exchange coupling in magnetic multilayers and sandwiches. A historical overview is given and a key advantage of MBE over alternative preparation techniques is emphasized: the ability to probe *in situ* the growth and structure of these materials. Recent work on surfactant-mediated growth of Co/Cu(111) sandwiches and the resulting reduction of growth-induced defects are discussed. A comparison of results for multilayers grown via MBE and sputtering is made for several GMR materials systems. It is seen that these preparation techniques are complementary.

Historical introduction

The technique of MBE was introduced in 1968 for the growth of thin films of III–V compound semiconductors (initially GaAs and $Al_xGa_{1-x}As$). It combined, for the first time, precise control, to atomic monolayer dimensions, of film thickness and composition profiles with the ability to study film growth in real time using a variety of *in situ* electron-beam probes. This improved definition of interfaces and dopant profiles led to significant improvements in the performance of conventional electronic devices (e.g., III–V quantum-well lasers and GaAs field-effect transistors) and led directly to new devices such as the modulation-doped field-effect transistor (also known as the high-electron-mobility transistor). Furthermore, MBE growth of undoped-GaAs/n-type $Al_xGa_{1-x}As$ structures led directly to the discovery of new and unexpected phenomena such as the fractional quantized Hall effect in a two-dimensional electron gas confined at the $GaAs/Al_xGa_{1-x}As$ interface. These developments had a profound influence on semiconductor physics and are reviewed elsewhere [1].

In the late 1970s MBE was applied to metal epitaxy, magnetic metal epitaxy, and eventually, in 1986 [2], to preparation of high-structural-quality, epitaxial magnetic rare-earth superlattices. The driving force for this new

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research direction was the expectation that, by analogy with semiconductor film growth, MBE could provide high-perfection, epitaxial, magnetic metallic structures which might exhibit new magnetic phenomena. This expectation was in fact realized by several discoveries in the late 1980s, including that of giant magnetoresistance. These are the subject of this review.

Here, it is worth pointing out some of the key features of MBE which distinguish it from the more conventional and widespread technique of sputtering for magnetic metal film preparation. In MBE, the substrate is maintained in ultra-high vacuum (UHV, i.e., $<10^{-9}$ mB) during its *in situ* preparation prior to epitaxy and during the growth process. The source of atoms or molecules for growth is the vapor flux from thermal sources, typically crucible sources (effusion cells) or electron-beam-heated metal charges. This UHV environment permits the substrate and growing film to be probed by a variety of electron-beam techniques to characterize their structures [by reflection, high-energy electron diffraction (RHEED), low-energy electron diffraction (LEED)] or compositions (Auger electron spectroscopy, X-ray photoelectron spectroscopy). UHV deposition minimizes incorporation of impurities into the film from background species. By contrast, in sputtering the substrate and targets are immersed in a gas (typically argon) at a pressure of a few mB. A high-voltage plasma discharge near the targets ejects a vapor flux toward the substrate. Thermalization of the ejected, energetic vapor species, by collision with gas atoms, takes place, and the effective energy of the arriving metal atoms at the substrate is comparable with that in MBE. In most sputtering systems in the 1980s, the growth chamber was not pumped to a UHV background, precluding effective substrate cleaning prior to film growth. In addition, impurities in the sputtering gas and targets were incorporated into the growing film. Thus, it is not surprising that high-perfection epitaxial metal films were not obtained by sputtering until the late 1980s after sputtering systems incorporated UHV design features previously found only in MBE systems.

The first high-perfection epitaxial magnetic superlattices were prepared in 1986, using MBE [2]. These rare-earth Gd/Y and Dy/Y superlattices provided an elegant demonstration of indirect exchange coupling of Gd (Dy) through the nonmagnetic Y layers via the RKKY (Ruderman–Kittel–Kasuya–Yoshida) interaction [2(a)–(c)]. This interaction describes indirect exchange coupling between isolated spins mediated by spin-polarization of the conduction electrons of the metallic host. In the rare-earth superlattices the coupling occurs (at temperatures below 150 K) via a helical spin-density wave in the Y. The demonstration of RKKY coupling in rare-earth superlattices led to interest in magnetic superlattices and sandwiches incorporating 3*d* transition metals with the prospect of discovering new *room-temperature* magnetic phenomena. This expectation was realized in the late 1980s. MBE was used to prepare the artificially layered magnetic structures in which antiferromagnetic interlayer exchange coupling [3, 4], enhanced magnetoresistance [5], and GMR [6] were discovered. The motivation for using MBE was the relative ease with which single-crystalline magnetic sandwiches and multilayers could be prepared by techniques developed earlier for epitaxial magnetic films [7–9] and structurally characterized *in situ*. For example, Fe epitaxy on GaAs(001) was used by Grünberg et al. [4] to seed the growth of (001)-oriented Fe/Cr/Fe sandwiches and by Baibich et al. [6] for (001)-oriented Fe/Cr multilayers. Single-crystalline (001)- and (110)-oriented structures with well-defined in-plane symmetry of magnetic and elastic properties made it easier to interpret the light scattering and magneto-optical data used by Grünberg et al. to demonstrate AF interlayer coupling in Fe/Cr/Fe sandwiches. However, another motivation was the assumption [4] that films of "reasonably good monocrystalline quality" were necessary for antiferromagnetic interlayer exchange coupling through Cr.

Subsequently, in 1991, Parkin et al. [10] (using a system incorporating UHV design features) discovered that magnetron-sputtered polycrystalline multilayers (Fe/Cr, Co/Cr, Co/Ru. . .) exhibited interlayer exchange coupling which oscillated from AF (antiferromagnetic) to FM (ferromagnetic) as a function of the nonmagnetic spacer thickness. Moreover, the magnetoresistance was oscillatory and its magnitude comparable with that in epitaxial structures (e.g., in Fe/Cr multilayers) prepared by MBE [6]. This discovery had several major implications. It showed that polycrystalline magnetic multilayers, prepared by the widespread technique of sputtering, had properties similar to those of single-crystalline multilayers prepared by MBE. This had technological significance, since sputtering is a manufacturing technique used for producing magnetic storage devices. It also raised questions of the influence of crystalline orientation, interface roughness, and structural quality of the multilayers on interlayer coupling and GMR. This stimulated widespread research on this topic, including *in situ* studies of multilayer growth and interface formation, for which MBE is particularly well suited.

A key example of *in situ* probing of magnetic film growth is the work of Unguris et al. [11, 12] on Fe/Cr/Fe sandwiches, which led to the discovery of short-period (about two monolayers) oscillations in interlayer exchange coupling as a function of Cr spacer thickness. Unguris et al. [11, 12] used UHV evaporation of Fe and Cr onto (001) facets of vapor-grown Fe whisker single crystals to form the trilayers. These facets are known from *in situ* STM (scanning tunneling microscopy) studies [13] to be atomically flat, with terraces as wide as 1 μm separating monoatomic steps. Such facets form an ideal substrate for epitaxial Fe/Cr/Fe trilayers, since Cr and Fe are bcc crystals with a lattice misfit of only 0.7%. The UHV growth environment permitted the use of two types of electron-beam probe to examine the trilayers *in situ*. First, RHEED (reflection high-energy electron diffraction) was used to probe the thickness of a Cr-wedged spacer layer grown onto the Fe facet. It is well known [14] that the specular beam intensity in RHEED oscillates in intensity with a period of a monolayer as the thickness increases during growth. By scanning the electron beam along the surface of the Cr wedge, the Cr thickness was determined precisely.

Figure 1 (upper trace) shows these oscillations. The second *in situ* electron-beam probe used was SEMPA (scanning secondary-electron microscopy with polarization analysis). SEMPA was used to probe the magnetic polarization of the underlying Fe facet, the Cr wedge, and the Fe film grown onto the Cr spacer. The spin polarization of the secondary electrons from the bare Cr, before and after removing the background polarization from the Fe substrate, is shown in the figure. The high polarization of electrons from the Fe at the start of the wedge decreased exponentially as the Fe electrons were attenuated by the Cr film of increasing thickness. From the exponential decay, a $1/e$ sampling depth for SEMPA in Cr was estimated to be only 5.5 ± 0.5 nm. The Cr polarization [P(Cr)], after removing the background polarization from the Fe substrate (see the third trace from the top in the figure), reversed nearly every other Cr layer. After this Cr wedge was coated with five monolayers of Fe, the SEMPA measurement revealed a spin polarization of the Fe [P(Fe)], opposite to Cr and with a period close to two monolayers of Cr. This showed that the Fe–Fe interlayer exchange coupling through Cr had this period. In fact, the phase slips seen in P(Cr) and P(Fe) at 22–25, 44–45, and 64–65 monolayers were consistent with a period of 2.11 ± 0.03 Cr monolayers. This discovery of short-period coupling oscillations was made possible by the extreme flatness of the substrate combined with sensitive, *in situ* RHEED and SEMPA probes, and the fact that interfaces between single-crystalline metals can have greater sharpness than those between polycrystals. Polycrystalline interfaces have an intrinsic roughness originating from differences in monolayer step height for different growth orientations. In order to detect the short-period coupling, Unguris et al. showed that the growth mode of Cr/Fe had to be strictly monolayer by monolayer. Scanning tunneling microscopy [12] revealed that this was the case for Cr grown onto the Fe whisker held at 300–350°C, but for growth at 100°C, the Cr growth front extended over at least four monolayers, and only the long-period coupling was present.

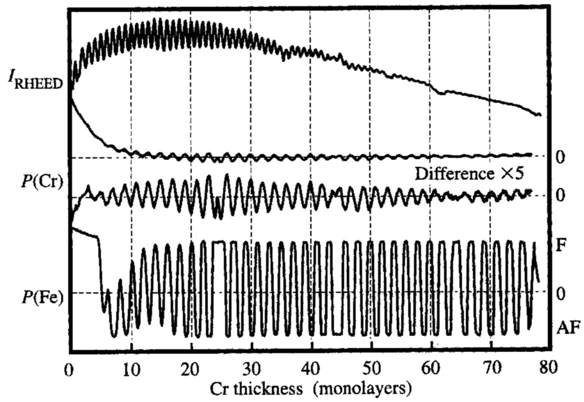

Figure 1

Scanned RHEED and SEMPA measurements from the same Cr wedge, showing the thickness dependence of the RHEED intensity oscillations (top), the spin-polarization dependence of secondary electrons emitted from the bare Cr [P(Cr)] before and after removing the Fe substrate polarization (middle), and the component of in-plane polarization (along the thickness wedge) from an Fe layer [P(Fe)] deposited on top of the Cr (bottom). F and AF respectively denote the ferromagnetic and antiferromagnetic states. From [12], with permission.

Following the discoveries of long-period interlayer exchange coupling and oscillatory GMR, the field widened with studies of many different combinations of materials. Parkin showed [15] that many spacer materials, in polycrystalline multilayers, had a similar long period (~10–12 Å) for coupling and oscillatory GMR. Studies of single-crystalline, (001)-oriented sandwiches, using thickness wedges of the spacer, showed that both short and long periods were present for Fe/Cr/Fe [16], Fe/Ag/Fe [17], and Co/Cu/Co [18]. Bruno and Chappert [19, 20] and Coehoorn [21] showed that these periods are consistent with a theory based on RKKY coupling. Essentially, a spin-density wave in the spacer layer has periods determined by stationary spanning vectors of the Fermi surface. Oscillatory interlayer exchange coupling can also be viewed [22] as a consequence of quantum confinement of electrons in the spacer layer. This viewpoint is discussed in the paper by Allenspach and Weber [23] in this issue. Experimental and theoretical aspects of these phenomena are covered in the paper by Nesbet in this issue [24]. In the present paper, we discuss a key issue which arose in the comparison between MBE and sputtered Co/Cu multilayers, namely the absence of oscillations in GMR or interlayer exchange coupling with spacer thickness for MBE-grown, (111)-oriented multilayers. This contrasted with the clear, long-period oscillations seen in magnetron-sputtered Co/Cu

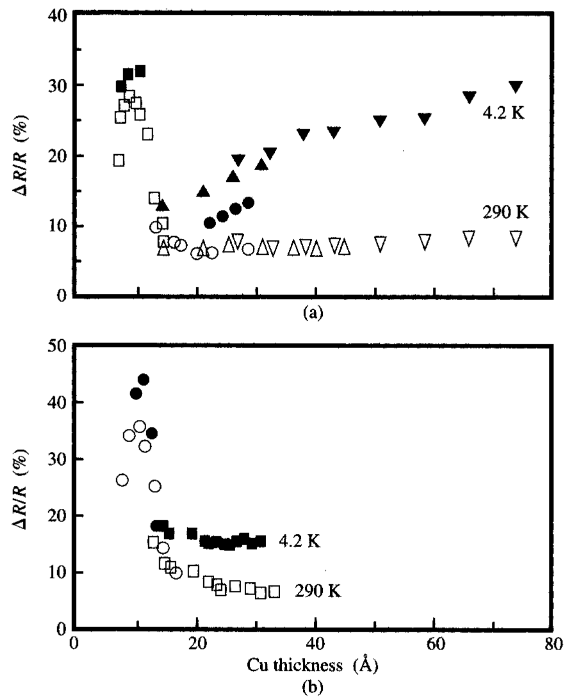

Figure 2

Magnetoresistance at room temperature (open symbols) and at 4.2 K (filled symbols) for Co/Cu(111) samples deposited at 0°C (a) and 150°C (b). Data are from a total of six wedge samples, with data from a single wedge plotted using the same symbol. The maximum applied field for each measurement was 60 kOe.

polycrystalline multilayers deposited on silicon substrates [15]. The *in situ* analysis techniques available in MBE allowed the growth of the multilayers to be explored in detail and the absence of oscillations to be related to growth-related defects. Following this discussion we review recent results on low-field GMR in permalloy (Ni_xFe_{1-x})/Au multilayers grown by MBE.

In situ studies of Co/Cu(111) multilayers and sandwiches

Co/Cu multilayers show [15(b), 25] some of the largest values of GMR for any materials system. Qualitatively, the reason for this is that, near the Fermi energy, the band structures of Co and Cu for majority carriers are similar, while the band structures for minority carriers are quite different [24]. This leads to a large contrast in spin-dependent scattering of majority and minority carriers as they cross the Co/Cu interfaces. Because of the large GMR values and related technological interest in this system, GMR and interlayer coupling have been studied extensively in Co/Cu multilayers and sandwiches using both magnetron sputtering and MBE as preparation techniques. For sputtered polycrystalline multilayers, it was found [15(b)] that for the most complete antiferromagnetic coupling between Co layers (corresponding to the largest GMR effect), it was necessary to use an Fe buffer layer to achieve flat, conformal Co and Cu layers. On the other hand, for structures prepared by MBE, it was found that while clear evidence existed for oscillatory exchange coupling through Cu in Co/Cu(001)-oriented sandwiches [26–29], evidence for oscillatory interlayer exchange coupling in Co/Cu(111)-oriented structures was contentious [30–32]. In fact, lack of complete antiferromagnetic coupling at the appropriate spacer thicknesses was reported by many groups [27, 33–38]. This lack of coupling was attributed [35] to structural defects such as local ferromagnetic bridges, although no direct structural data were available at the time to support this suggestion.

In situ studies of MBE-grown Co/Cu multilayers and sandwiches were then carried out to determine the effect of growth conditions on interlayer exchange coupling and GMR. Harp et al. [38] prepared Co/Cu(111)-wedged multilayers in which the Cu layers were nearly linear wedges in thickness but the Co layers were of constant thickness. These multilayers were grown onto highly oriented Cu seed films grown onto Pt(111)/sapphire(0001). Magnetoresistance measurements as a function of thickness for these multilayers are shown in **Figure 2**. Measurements at 4.2 K and 290 K revealed only a single peak in magnetoresistance at a Cu thickness of ~10 Å. Multilayers grown at 0°C and 150°C showed qualitatively similar behavior, but the magnetoresistance peak was significantly larger at 150°C. Magnetization data revealed that only a minor fraction of the sample had antiferromagnetic coupling through Cu for both growth temperatures. The *in situ* techniques of X-ray photoelectron diffraction (XPD) and X-ray photoelectron spectroscopy (XPS) were used to examine the growth modes of Co/Cu(111) and Cu/Co(111), since it was suspected that the origin of ferromagnetic coupling between Co in the multilayer was growth-induced. It was found that at 0°C the Co grew in a diffusion-limited layer-by-layer mode, with accumulated roughness as the multilayer was grown. On the other hand, for growth of Co/Cu(111) at 150°C, Co grew in a layer-by-layer mode with minimal accumulated roughness but with the surface segregation of a Cu monolayer.

A key finding from XPS measurements was that growth of Cu/Co and Co/Cu/Co at 0°C revealed no evidence of interdiffusion or Cu surface segregation. On the other hand, for growth at 150°C, significant interdiffusion was apparent from detection of Co at the surface of thick Cu films and of Cu at the surface of thick Co films. The diffusion of Co through thick (~100 Å) Cu films can be

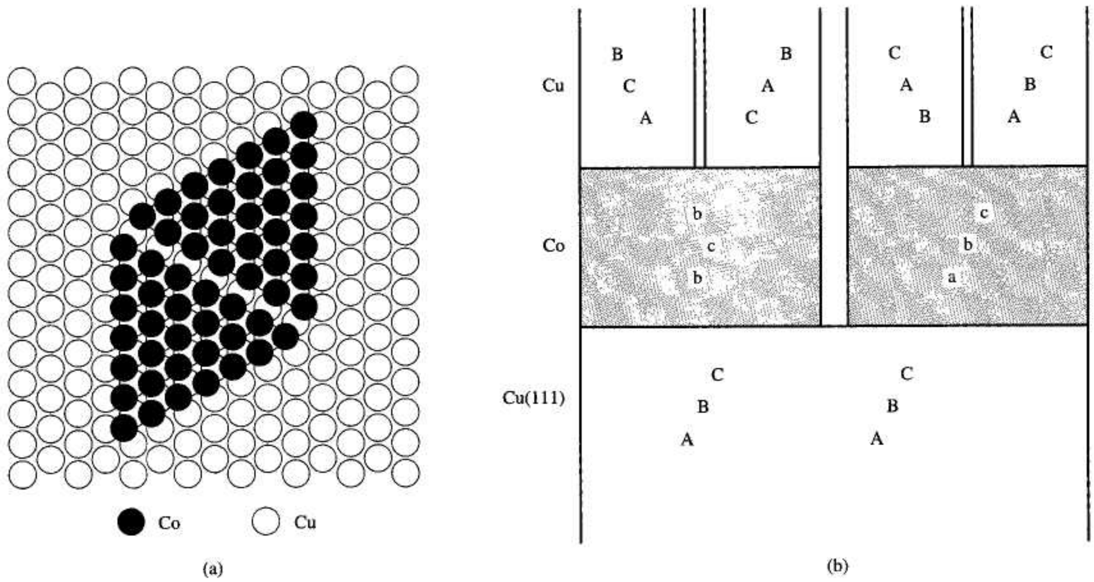

Figure 3

Schematic diagram showing (a) twinned islands on Cu(111) and (b) stacking sequence in Co/Cu bilayers. From [39], with permission.

attributed to preferential diffusion along crystalline defects, e.g., twin boundaries known to be present in the multilayer from RHEED and LEED as well as X-ray diffraction studies. Bulk diffusion of Co through Cu can be excluded as a mechanism at this temperature. For growth of Co/Cu(111) multilayers, there appears to be no optimal growth temperature to avoid growth-induced defects. At 0°C, the interfacial roughness is accumulative, and for growth at 150°C there is interdiffusion of Co through the Cu spacers consistent with ferromagnetic bridging of Cu.

A more detailed picture of Co/Cu(111) and Cu/Co/Cu(111) growth was developed from STM (scanning tunneling microscopy) studies [39]. The growth of Co/Cu/Co trilayers onto bulk single crystals of Cu(111) was examined by STM, and it was found that at 0°C, Co nucleated on Cu(111) in triangular islands which were two atoms thick. The islands were of two orientations related by a 180° twin orientation. **Figure 3(a)** shows a schematic diagram of twinned islands of Co on Cu(111). STM [39(a)] showed that the Co islands nucleated at each of the two threefold sites of the fcc Cu(111) face. As indicated in **Figure 3(b)**, one set of islands follows the correct fcc stacking sequence (ABCabc), while the other set has a stacking error at the interface (ABCbcb). The islands do not coalesce, but have a vertical channel at the interface. The measured [39(a)] widths of these channels, after deposition of five monolayers of Co at room temperature, varied widely, reaching ~30 Å in places. When Cu is nucleated on top of the twinned Co islands, it also develops twins (Figure 3) with vertical channels at the island interfaces. These channels persist to Cu overlayer thicknesses of many monolayers; if they become partially filled on subsequent deposition of Co, they can act as ferromagnetic bridges between Co layers, obscuring observation of indirect exchange coupling and causing a fraction of the sample to be ferromagnetically coupled regardless of whether oscillatory indirect exchange coupling is present. As Gradmann et al. [40] pointed out, incomplete coalescence of fcc metals, in heteroepitaxy on single-crystalline (111) metal surfaces, is well known from early observations of metal films using plan-view transmission electron microscopy studies. They showed that oscillatory exchange coupling could be observed with epitaxial Co/Cu/Co trilayer growth on highly oriented Cu(111) films grown onto sapphire $(11\bar{2}0)$ substrates if the Co thickness was restricted to one monolayer, and pointed out that the spatial separation of ferromagnetic bridges was an extrinsic feature which depends on the details of film growth. Camarero et al. [39(b)] subsequently showed that the twinning of Co/Cu(111) could be inhibited through the use of Pb as a surfactant. If the Co was deposited onto a Pb monolayer on Cu(111), LEED I–V studies showed that the Co islands grew in a single

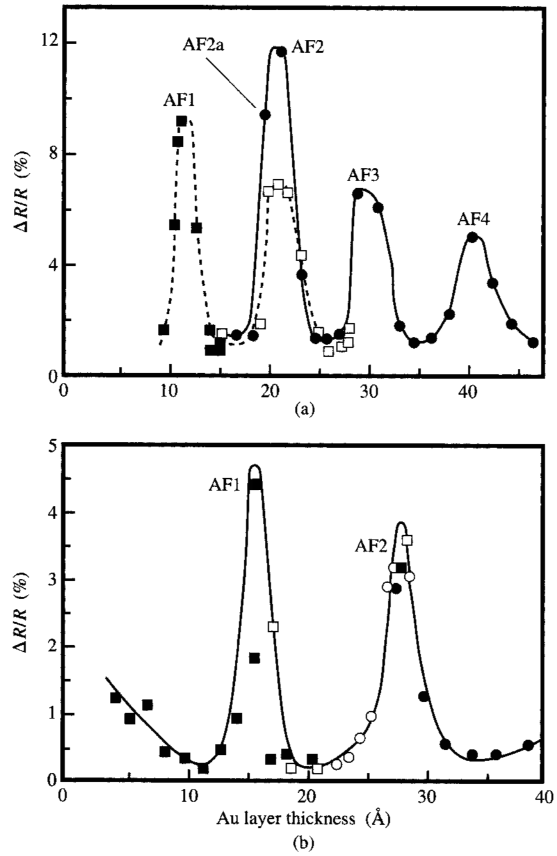

Figure 4

(a) Oscillatory behavior of magnetoresistance ($\Delta R/R$), measured at 298 K, as a function of Au spacer thickness in Py/Au(111) and $Py_{86}Au_{14}$/Au(111) multilayers prepared by MBE. The closed circles represent data for Py/Au multilayers; the open and closed squares represent data for $Py_{86}Au_{14}$ multilayers. The Py–Au and Py thicknesses were ~30 Å. From [48], with permission. (b) Oscillatory behavior of magnetoresistance ($\Delta R/R$, measured at 298 K, as a function of Au spacer thickness in polycrystalline Py/Au multilayers for four series of Py/Au multilayers, 40 Å Cr/Cr/[15 Å Py/Au]$_{11}$/15 Å Py/10 Å Cr. The solid and open squares correspond to samples deposited on dynasil, and the solid and open circles are samples deposited on silicon. From [49], with permission.

orientation with suppression of stacking faults at the Cu/Co interface. Cu/Co/Cu trilayers thus grew untwinned with the Pb monolayer floating to the top of the structure.

Recently, Camarero et al. [41] closed the structure-property "loop" for this particular materials system by showing that untwinned Co/Cu/Co trilayers, prepared with surfactant Pb, indeed showed (nearly) complete antiferromagnetic coupling at a Cu thickness of four monolayers. They used intensity oscillations in reflection electron diffraction to show that Pb induced a layer-by-layer growth mode of Co at room temperature and used the magneto-optical Kerr effect to demonstrate complete AF coupling. Independently, Egelhoff et al. [42] showed that use of Pb as a surfactant in preparation of polycrystalline Co/Cu spin valves improved their GMR behavior. The use of Au and In as surfactants in preparation of spin-valve structures was also investigated [42].

Comparison between MBE growth and sputtering of GMR and AF coupled structures

As discussed above, the absence of oscillatory GMR and interlayer exchange coupling in MBE-grown Co/Cu(111) multilayers was correlated with growth-induced defects arising from twinned islanding of Co on Cu(111) terraces. Polycrystalline, sputtered Co/Cu multilayers are likely to have a very small fraction of (111) terraces available during growth, and if their size is small enough, migration of adatoms to surface steps precludes islanding. For this reason, the mechanism for ferromagnetic bridging, operative in MBE growth of Co/Cu(111), is absent. This explains the clear observation of long-period, oscillatory GMR and interlayer coupling for these sputtered structures. However, it raises the question as to whether ferromagnetic bridging is present in *epitaxial* Co/Cu(111) multilayers prepared by sputtering. In recent years it has been shown that growth of highly oriented and single-crystalline magnetic structures by sputtering is possible if epitaxial seed films are first grown onto single-crystal substrates. For example, by using seed-film/substrate combinations similar to those in MBE, highly oriented or single-crystalline magnetic metal sandwiches and multilayers can be grown by sputtering [43, 44]. Quantitative comparison of structural quality is difficult because the mosaic spread of the substrate is a variable factor. X-ray rocking curve widths provide a measure of mosaic spread, and Harp and Parkin [43] report a FWHM of 1.7° for the (200) superlattice peak from a sputtered epitaxial Co/Cu(100) structure comprising Pd 30 Å/[Cu 7 Å/Co 8 Å]$_{40}$/Pd 50 Å/MgO(100). This is larger than but comparable with rocking curve widths for the (111) superlattice peak in MBE-grown Co/Cu multilayers grown on sapphire [45]. Recently, Parkin [25] has prepared epitaxial Co/Cu multilayers by sputtering with [001], [110], and [111] growth axes. Interestingly, the second AF maximum and higher-order maxima are least well defined for the [111] growth orientation. This suggests that the same type of growth defects as in MBE Co/Cu(111) structures are present in the sputtered structures. Also according to Parkin [25], multilayers with the [001] and [110] orientations show clear, long-period oscillations and evidence for short-period oscillations. In addition, the magnitude of GMR at the first AF maximum is orientation-independent.

The clearest picture to date of oscillatory interlayer coupling through Cu(001), in the Co/Cu(001) system, comes from the work of Weber et al. [23, 46]. They have shown that MBE growth of wedged, single-crystalline Co/Cu/Co(100) trilayers on single-crystal Cu(001) substrates, combined with "spin-SEM" (identical with SEMPA), provides a detailed picture of both long- and short-period oscillatory interlayer exchange coupling. The measured long and short periods are in good agreement with theory. However, the relative amplitudes of the two types of oscillations changed dramatically when a different Cu substrate crystal was used. This was attributed to the influence of substrate on Cu spacer layer roughness, demonstrating how sensitive interlayer coupling is to growth conditions and interface roughness. Indeed, magnetic anisotropy is also sensitive to roughness on the atomic scale, for this material system [46]. Monolayer periods of roughening and in-plane lattice spacing [47] have been detected using RHEED observations of Co/Cu(001) growth in real time, demonstrating the value of *in situ* probes for investigating oscillations in magnetic anisotropy. This emphasizes an advantage of MBE over sputtering: The background pressure during magnetron sputtering precludes the use of electron beam probes such as RHEED and SEMPA *during* growth.

We have seen that, for Co/Cu(111) multilayers, MBE growth results in structures which are dramatically influenced by growth-induced, ferromagnetic bridges. This is in contrast with the case of permalloy (Ni_xFe_{1-x})/Au(111) multilayers grown by MBE, which show [**Figure 4(a)**] a clearly resolved, long-period (~10 Å) oscillation in GMR and interlayer exchange coupling extending out to the fourth AF coupling maximum at ~40 Å [48]. Sputtered, polycrystalline permalloy/Au multilayers, with the same number of periods, also exhibit [49] oscillatory GMR and exchange coupling [**Figure 4(b)**], but there are significant differences in magnetic behavior from the MBE-grown multilayers. For example, the maxima in magnetoresistance for the MBE-grown multilayers are larger: 9% vs. 6% (AF1) and 11.6% vs. 3.8% (AF2). Other differences are weaker interlayer exchange coupling and a larger oscillation period (~12.5 Å) for the sputtered samples. These differences probably reflect structural differences, including texture and length scale of interface roughness [48, 49] of the multilayers. For both MBE and sputtered multilayers, the coupling strength through Au(111) is much weaker than for coupling through Cu(111), and the multilayers exhibit large changes in resistance per unit field: ~1% per Oe for AF2 (sputtering) and AF4 (MBE). These are the largest room-temperature magnetic field sensitivities yet reported in simple magnetic multilayers.

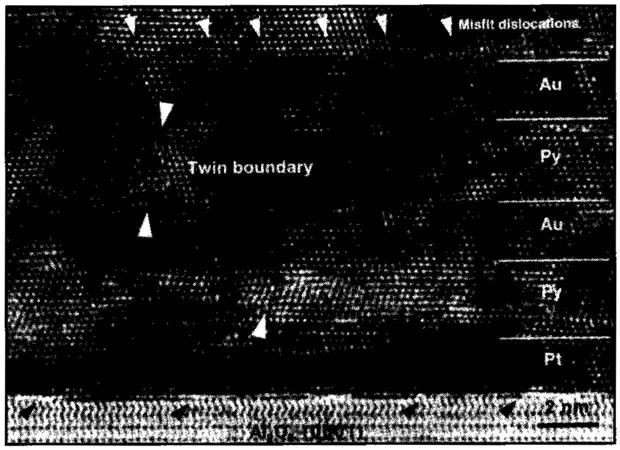

Figure 5

High-resolution cross-sectional image of Py/Au(111) multilayer [AF2a: see Figure 4(a)] for the as-grown sample. The first five layers and the Pt seed layer are visible. Steps at the Pt/sapphire (0001) interface are indicated, as well as twin boundaries and misfit dislocations in the Py layers. From [50], with permission.

It is interesting that the MBE-grown permalloy/Au multilayers have complete AF coupling, at AF2 for example, in contrast with MBE-grown Co/Cu(111) multilayers where the AF-coupled fraction is the minority, even at AF2. Permalloy/Au multilayers are also twinned, however; high-resolution, cross-section transmission electron microscopy (HRXTEM) studies [50, 51] reveal that in as-grown permalloy/Au multilayers (with the Au thickness at AF2: ~21 Å) the twin boundaries, parallel to the growth direction, originate from the Pt seed film but do not extend through the multilayer stack. They are confined to the first one to three layers of the multilayer. In addition, twin boundaries in the permalloy layers tend to terminate at the interface with the adjacent Au layers. This twin-termination behavior can be seen in **Figure 5**, which shows a HRXTEM micrograph of the first five layers in a permalloy/Au multilayer with the Au thickness (~21 Å) corresponding to AF2. Other localized defects such as misfit dislocations are also present in the structure; however, the twin-termination feature of growth may be the reason why ferromagnetic bridges are largely absent in these multilayers. Ferromagnetic bridging by diffusion along twin boundaries is limited. Our studies [52] of annealing of these multilayers show that widespread ferromagnetic bridging eventually occurs after annealing at 250°C by local alloying of permalloy with Au, leading to fluctuations in the Au spacer thickness and direct contacts between adjacent permalloy layers.

A direct comparison between epitaxial growth via sputtering and MBE of the same materials system can be made in the case of Fe/Cr multilayers. Fullerton et al. have shown [53] that epitaxial (001)-oriented multilayers of high structural quality can be prepared by dc magnetron sputtering using Cr seed films grown at high (600°C) substrate temperatures. These multilayers have coherence lengths along the growth direction of ~430 Å and a record value of GMR (~150% at 4.2 K) for a $[Fe(14 Å)/Cr(8 Å)]_{50}/Cr(100 Å)/MgO(001)$ structure—a value that is much higher than the GMR values (~60% at 4.2 K) reported in the initial discovery of GMR by Baibich et al. [6] in MBE structures. This demonstrates that, with optimization of growth and seeding techniques, sputtered epitaxial multilayers of structural quality at least comparable to MBE can be grown. Structural perfection, flatness, and surface impurity concentration of seed film and substrate are probably the key factors in controlling multilayer quality, rather than intrinsic differences in the deposition technique. The absence of any evidence of *short-period* oscillations of GMR or interlayer exchange coupling in these structures also shows that the interface roughness does not approach that of MBE-grown Fe/Cr(001)/Fe(001) sandwiches grown [11] onto single-crystalline Fe(001) whisker facets. In the latter case, the substrate terrace dimensions approach microns.

Discussion

The contributions of MBE and sputtering to this field continue to be complementary. *In situ* probing of MBE-grown, epitaxial Co/Cu/Co, Cr/Fe/Cr, and other sandwiches using RHEED for precise spacer thickness determination, combined with the magnetic probes of spin-SEM (SEMPA) and the magneto-optical Kerr effect, are leading to a detailed picture of both short- and long-period oscillatory interlayer exchange coupling. Oscillations in several other magnetic properties (magnetic anisotropy, magneto-optical response) also show [23] oscillations with interlayer thickness originating from intrinsic phenomena (quantum-well states, spin density wave oscillations). In some cases, extrinsic phenomena also play a role, as in the case of monolayer-period oscillations in magnetic anisotropy of Co/Cu(001) resulting from periodic changes in overlayer roughness. For some materials systems, especially Co/Cu(111)-oriented sandwiches and multilayers, the growth process can produce defects (local ferromagnetic bridges) which have a strong extrinsic influence on GMR and interlayer exchange coupling. These defects result from incomplete coalescence of twinned islands of Co and Cu and can be suppressed by growth in the presence of a surfactant (Pb) which eliminates twinning. On the other hand, in permalloy/Au(111) superlattices, twinning is also present, but there is little evidence for ferromagnetic bridging. High-resolution electron microscopy studies suggest that this may be due to the tendency for twin boundaries in the permalloy to terminate at the interface to the Au layers. The mechanism for this behavior is not yet clear, and further work is needed to examine growth mechanisms and step edge barriers to adatom migration. Similarly, in the Co/Cu(001) system, oscillatory interlayer exchange coupling is sensitive to the growth process in that the relative amplitudes of short- and long-period interlayer exchange coupling are influenced by initial roughness of the substrate and of the Cu/Co interfaces. Here also, STM studies of the development of interface roughness as a function of growth conditions, combined with *in situ* probes of the coupling, are needed.

Summary

A major role of MBE in connection with GMR and interlayer exchange coupling is in developing a more complete understanding of these phenomena and in clarifying the influence of growth conditions on them. SEMPA-wedge experiments and microstructural studies of MBE-grown permalloy/Au(111) multilayers exemplify this approach. On the other hand, sputtering (by dc magnetron or ion-beam methods) is the deposition method of choice for magnetic storage devices (for example, spin-valve GMR read heads) because of its high throughput. Sputtering can be used to produce magnetic sandwiches and multilayers with record values of GMR. Its use has facilitated major contributions to our understanding of GMR and interlayer exchange coupling as well as an expansion of the range of materials systems exhibiting these phenomena. MBE growth and sputtering can therefore be viewed as complementary techniques.

Acknowledgment

I acknowledge, with thanks, helpful discussions with Ronald F. Marks, and his assistance in preparation of this manuscript. The author is particularly grateful to N. Thangaraj and Kannan M. Krishnan for recording and analysis of the high-resolution electron microscope image reproduced in this paper.

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Received January 9, 1997; accepted for publication July 15, 1997

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