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Roughness-induced coupling between ferromagnetic films across an amorphous spacer layer

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The exchange coupling between an amorphous ferromagnet and a polycrystalline Fe layer across an amorphous AuSn spacer is studied by spin-polarized secondary-electron emission. No antiferromagnetic or oscillatory exchange coupling is observed. Instead, we find a coupling of unexpectedly long range which favors perpendicular alignment of the ferromagnets at larger spacer thicknesses. At a spacer thickness of 30 to 50 Å the magnetization direction of the top Fe layer continuously changes from ferromagnetic to 90° alignment and remains in its perpendicular orientation for AuSn films up to at least 100 Å. The rather weak thickness dependence of the observed coupling points at a recently proposed magnetic dipole interaction as possible mechanism. Following models by Demokritov *et al.* and by Néel and using the roughness parameters extracted from scanning tunneling microscopy we calculate the coupling strengths of the dipolar interactions. The experimental results can be described with the models. We confirm that in structurally disordered multilayers roughness-induced magnetic dipolar interactions can cause biquadratic as well as bilinear coupling, while the quantum-mechanical exchange is strongly suppressed. The calculated coupling strengths set an upper bound of 0.002 erg/cm^2 for the quantum-mechanical exchange across this disordered metallic spacer. [S0163-1829(97)04317-8]

I. INTRODUCTION

The discovery of oscillatory exchange coupling between ferromagnetic layers separated by a nonmagnetic spacer[1–3] has triggered a rush of experimental and theoretical investigations. Various studies established that oscillatory exchange coupling is a general phenomenon for most transition-metal and noble-metal spacers.[4] Further progress was achieved by preparing high-quality epitaxial trilayers with wedge-shaped spacers, where the coupling exhibits multiperiodic behavior.[5,6] Along with these experimental findings several theoretical approaches have been proposed to explain the physical origin of oscillatory exchange coupling.[7,8] In metallic multilayers the exchange interaction is propagated by conduction electrons and the different existing theories are obtained as limiting cases of the same underlying physics[9] starting from different points. The most common description is a Rudermann-Kittel-Kasuya-Yosida-type (RKKY) coupling modified by taking into account the discreteness of the spacer material.[7] In another picture[8] quantum wells produced by the different spin states of the electrons in the ferromagnets can explain the oscillatory behavior of the coupling. In both cases the exchange is strongly related to the topology of the Fermi surface and the discreteness or the periodicity of the spacer material. These basic principles have recently been confirmed by *ab initio* calculations.[10] Furthermore it has been discovered[11] that in the transition region between ferromagnetic and antiferromagnetic coupling the magnetization of the two ferromagnetic films favors perpendicular orientation which usually is parametrized in a biquadratic expression. First this biquadratic coupling only appeared at the crossovers where the oscillatory term vanishes. Recent observations have shown that this type of coupling can be of considerable strength with antiferromagnetic spacers,[12] and it can—mainly at low temperatures—even dominate the exchange interactions in the presence of interface roughness.[13] Various models based on exchange have been put forward to describe the biquadratic coupling.[14–19] Moreover, a magnetic dipole mechanism arising from interfacial roughness and resulting in long range biquadratic coupling has recently been proposed by Demokritov *et al.*,[20] and experimentally confirmed by the same group.[13] In the present study we will provide further evidence of the importance of this dipolar coupling mechanism.

Another issue of interest is the role of disorder within the bulk of the spacer material. The surprising observation of oscillatory exchange coupling across amorphous semiconductors by Toscano *et al.*[21] has indicated that oscillatory exchange coupling even exists in the case of nonmetallic as well as noncrystalline spacer materials. This calls for an extension of the established theories of the exchange coupling. Substitutional alloys, furthermore, offer the possibility to alter the electronic structure by varying the composition and thus to analyze the relationship between the coupling and the electronic structure. This kind of ‘‘Fermi-surface engineering’’ has been carried out in several experiments on crystalline Cu alloys with different substituents,[22–25] and the results are understood in the frame of RKKY coupling.[22,26] Beyond that, the influence of substitutional disorder at the interfaces and in the bulk of the spacer has been studied from first principles.[27,28] However, magnetic interactions in *amorphous* alloy multilayers have not been explored experimentally up to now. From a theoretical point of view the definition of a spherelike Fermi surface is still possible[29] and an RKKY-like coupling mediated in the free-electron gas by spin-density waves seems to be imaginable although the structural periodicity is absent in the amorphous state. In fact, predictions for oscillatory exchange coupling exist even in the limit of a free-electron approximation.[7,30] Therefore amorphous multilayers seem to be a good candidate to question the existing theories.

Along this line of thought we have set out to investigate magnetic interactions between two ferromagnets across an amorphous metallic AuSn alloy spacer. Surprisingly we find magnetic coupling to exist which is of unexpectedly long range and which favors perpendicular alignment of the ferromagnetic layers at larger spacer thicknesses. No antiferromagnetic or oscillatory behavior of the coupling is observed. At a spacer thickness of 30 to 50 Å the coupling changes from ferromagnetic to 90° alignment and remains in this state for spacer thicknesses of up to 100 Å. Structural analysis of the substrate by scanning electron microscopy (SEM) and scanning tunnel microscopy (STM) has revealed a certain surface roughness which can give rise to magnetic dipole fields outside the ferromagnetic layer and result in magnetostatic coupling. We confirm that these roughness-induced dipolar interactions can play a decisive role and even dominate the coupling. The results are quantitatively discussed in terms of magnetic dipole interactions following the models by Demokritov *et al.*[20] and by Néel.[31] From these calculations we can estimate an upper bound of about 0.002 erg/cm^2 for the strength of a RKKY-like coupling in the present amorphous alloy.

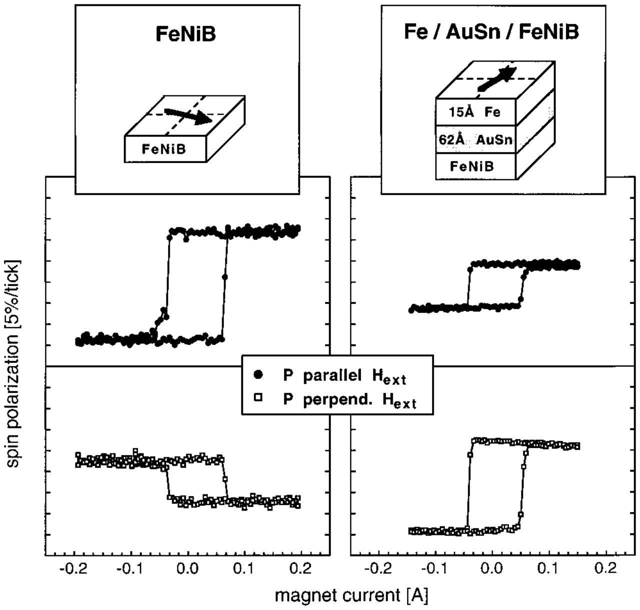

FIG. 1. SPSEE hysteresis loops of the FeNiB substrate (left panels) and of a 90° coupled Fe/AuSn/FeNiB trilayer (right panels) at 40 K. The nominal AuSn spacer thickness is 62 Å. Illustrated are the in plane components of the spin polarization parallel (top panels) and perpendicular (bottom panels) to the external field H_{ext}.

II. EXPERIMENTAL

As an experimental tool for magnetometry we utilize spin-polarized secondary electron emission (SPSEE).[32] In this technique the sample surface is irradiated by an unpolarized primary electron beam of 1 keV. A cascade mechanism generates low-energy secondary electrons of high intensity and, in a lucky combination, of high polarization. The surface normal emission of secondary electrons is subsequently submitted to a 100-keV Mott detector for spin analysis. The polarization is defined as $P=(N\uparrow - N\downarrow)/(N\uparrow + N\downarrow)$, where $N\uparrow$ $(N\downarrow)$ is the number of electrons parallel (antiparallel) to the quantization axis of the spin detector. The chosen arrangement of the detector enables us to measure the two in-plane components of the spin polarization which is proportional to the magnetization in a surface region of 4–5 Å.[33,34] This high surface sensitivity allows to directly probe the magnetization of the outermost layer and therefore makes SPSEE a good instrument for *in situ* analysis of multilayer systems. We monitor the response of an exchange-coupled surface layer and consequently study the coupling itself. As a ferromagnetic substrate we use an amorphous FeNiB alloy ribbon with a low coercivity, mounted onto a horseshoe magnet. It is magnetized in plane and exhibits a magnetic easy direction which lies roughly parallel to one quantization axis of the detector.

As an amorphous spacer we use AuSn. We have chosen a AuSn alloy because the structure of noble-metal tin alloys has been studied very intensely in the past and their electrical properties are well known.[28,35–37] Among noble-metal compounds AuSn exhibits the greatest stability and the temperature dependence of its transport properties is almost reversible below the crystallization temperature of at least 250 K.[35,37] Vapor-quenched AuSn alloys in the concentration range from 20 to 80 at. % grow amorphously on glass substrates held at 77 K or below.[35–38] For that reason we prepare the AuSn layers at 40 K by coevaporation from *e*-beam sources onto the *in situ* sputter-cleaned FeNiB metglass surface. We prepare Au-rich samples with a noble-metal content of about 66 at. %. The deposition rates of Au and Sn are individually checked by a quartz thickness monitor. Additionally cleanliness and composition are verified by Auger electron spectroscopy. The thickness is determined by the relative changes of the Fe $L_3M_{45}M_{45}$, the Au $N_5N_7O_4$, and the Sn $M_5M_{45}M_{45}$ Auger intensities upon evaporation.[39] From the exponential variation of the Auger intensities with increasing layer thickness we conclude that the amorphous AuSn films grow homogeneously. The trilayer is completed by deposing a 15-Å-thick Fe layer at the top. All preparations and measurements are performed at 40 K under UHV conditions with a base pressure of 10^{-10} Torr.

III. RESULTS

The magnetic properties of the trilayer at fixed temperature are determined by measuring the SPSEE hysteresis loops of the top layer. The results of a trilayer with a 62-Å-thick AuSn spacer is presented in Fig. 1, right panels. The corresponding hysteresis loops observed before on the substrate are illustrated in the left panels. Depicted are the in-plane components of the polarization parallel (upper panels) and perpendicular (lower panels) to the external field. The top Fe layer shows a square hysteresis loop with the same small coercive field as observed on the substrate. This unambiguously indicates nonzero magnetic coupling between the top layer and the substrate. Surprisingly the perpendicular component of the top layer has become significantly larger than the parallel one. The magnetization of the top Fe layer is at right angle with the one of the substrate. As a first main result the coupling angle, i.e., the angle between the magnetizations of the substrate and of the top layer, respectively, as a function of the spacer layer thickness is presented in Fig. 2, top panel. Up to 30 Å spacer thickness the Fe film

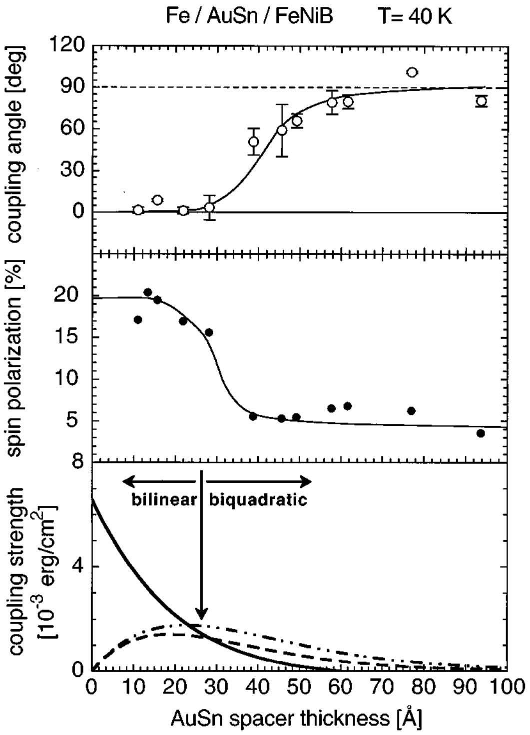

FIG. 2. Comparison of the coupling angle (top panel), remanent spin polarization (center panel), and estimated dipolar coupling strengths (bottom panel) versus interlayer thickness. The lines in the bottom panel represent bilinear (solid line) and biquadratic (dashed and dashed-dotted line) coupling as described in the text.

couples ferromagnetically to the substrate. Going to thicker interlayers the magnetization continuously changes its orientation from 0 to 90°. From 50 to at least 100 Å the coupling favors perpendicular alignment of the ferromagnets. No antiferromagnetic coupling or oscillatory behavior is found.

In the case of nonferromagnetic alignment the coupling strength can be determined by measuring the external field at which the intrinsic coupling is compensated. The high sensitivity of low-energy electrons to external fields, however, makes this measurement quite unreliable. As an alternative, in the case of disordered magnetic films without any preferred magnetization direction and hence without remanence, the coupling strength is reflected by the induced absolute polarization of the top layer. Assuming a linear dependence of the magnetization of the top layer on the exchange field far from saturation, the measured remanent polarization gives a good qualitative picture of the coupling strength for sufficiently weak coupling. Figure 2, center panel, shows the total remanent spin polarization versus interlayer thickness. Each data point represents an average over the full width of the sample. Up to about 30 Å the top layer magnetization is saturated. For thicker spacer films the coupling is significantly reduced, but remains nearly constant and shows no remarkable thickness dependence. We note that the absolute magnetization data exhibit some scatter over the whole width of the sample, while the coupling angle is nearly unchanged.

IV. DISCUSSION

In the full thickness range we do not find antiferromagnetic coupling or oscillatory behavior. From this we conclude that an RKKY coupling across amorphous AuSn does either not exist or it is weaker than and covered up by the dipolar effects discussed below. Generally, coupling across crystalline noble-metal spacers is reported to be rather weak.[40,41] First experimental studies failed in detecting coupling in polycrystalline Cu, Ag, and Au multilayers.[4] Interface roughness as well as alloying in epitaxial or strongly textured samples likewise tend to reduce the coupling strengths.[23,25] The lack of oscillatory exchange coupling across this amorphous spacer material now further supports the accepted models for metallic multilayers which are based on the periodicity of the spacer and the existence of a well defined Fermi surface. Disorder alters the electronic structure of the spacer material and the singularity in the occupancy near the Fermi level is smeared out even at low temperatures. This Fermi surface ‘‘dusting’’ most likely suppresses an RKKY-like coupling across amorphous spacers. On the other hand, the present results shed some light on the previously reported exchange coupling through amorphous semiconductors.[21] The findings suggest that the exchange across amorphous semiconductors, where the electronic structure is determined by localized defect states, presumably is of quite different origin.

The main observation is the existence of pronounced 90° coupling of unexpectedly long range. In most models which attempt to describe this so-called biquadratic coupling[14–19] an intrinsic type of mechanism, arising from the electronic structure of an ideal trilayer, is proposed. The resulting coupling strengths are very small compared to the so-called bilinear one, and decrease rather fast with increasing distance from the interface. Moreover, in these intrinsic models as well as in Slonczewsky's fluctuation mechanism[18] the biquadratic coupling is somehow proportional to a RKKY exchange. The present observations all are at variance with these properties. However, a magnetic dipole mechanism resulting in long-range biquadratic coupling has recently been proposed[20] and experimentally verified.[13] This interaction is based on magnetic dipole fields created by the interface roughness of the magnetic layers. In order to test whether roughness also in the present case is a possible cause of the observed coupling we have carried out structural investigations by scanning electron microscopy (SEM) and by scanning tunneling microscopy (STM). Straightforward SEM studies show that our amorphous Fe-Ni-B substrates exhibit no remarkable surface structure. Only under gracing incidence of the primary electrons, at an angle of 82–85° from the surface normal, a certain surface structure becomes visible. Smooth hills with a typical distance of a few 10 nm are clearly recognizable. For a quantitative analysis of the observed surface roughness we perform STM measurements under ultrahigh-vacuum conditions. A typical STM image of the FeNiB surface is presented in Fig. 3. The surface of the amorphous Fe-Ni-B samples is generally very flat, but composed of a network of small islands. The gray scale range of this 2000×2000 Å^2 image is only 7.9 Å. White and black areas indicate peaks and valleys, respectively. From statistical evaluations of several images we extract characteristic roughness parameters. The amorphous substrates exhibit an undulatory surface structure with typical peak to peak distances of 20 nm and peak heights of 5 Å. The structure does not remarkably alter upon ion sputtering, a fact which is

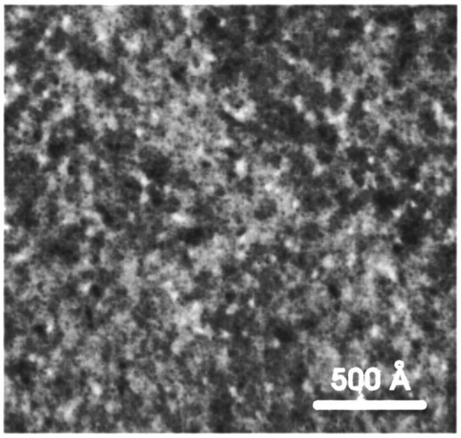

FIG. 3. STM topographic image of a sputtered FeNiB alloy surface. The gray scale range is 7.9 Å. A sample voltage of 0.4 V and a tunnel current of 0.5 nA are used to collect the image.

supported by similar STM studies on various amorphous ribbons.[42] We conclude that the observed surface roughness indeed can give rise to magnetostatic coupling effects.

V. MAGNETIC DIPOLE MECHANISMS

Generally, the coupling interaction between two ferromagnetic films across a nonmagnetic spacer layer can be expressed by the following phenomenological equation:

$$E=-J_1\cos\Theta-J_2\cos^2\Theta, \tag{1}$$

where E denotes the interlayer coupling energy per unit area, and Θ is the angle between the magnetization direction of the two films with respect to each other. J_1 and J_2 are the bilinear and biquadratic coupling terms, respectively.

Next we attempt to explain the observed interlayer coupling in terms of dipolar interactions by strictly applying the models by Demokritov *et al.*[20] and by Néel.[31] The roughness-induced dipole fields can cause biquadratic coupling and also contribute to the bilinear one. The model by Demokritov *et al.* is treated first because we wish to put some emphasis on the biquadratic term. We note, however, that the general idea of this model is based very much on Néel's original approach which yields bilinear coupling. We briefly describe the basic ideas behind the two models and present the results. For a detailed account we wish to refer to the original papers.

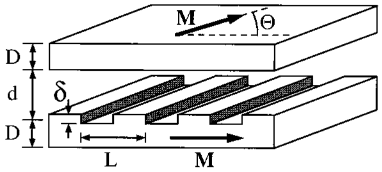

FIG. 4. Schematic illustration of Demokritov's model for biquadratic coupling (Ref. 20). Both magnetic films of thickness D separated by a spacer of thickness d are magnetized in plane. Θ is the angle between the top and bottom layer magnetizations. The bottom film has periodic interfacial terraces with period L and step height δ.

A perfectly flat ferromagnetic layer which is in-plane magnetized does not produce a magnetic field outside itself. On the other hand, in the presence of surface roughness the situation is different. Terraces at the interface of a film provoke a magnetic-dipole field outside the layer which decays exponentially[20] with increasing distance sufficiently far away from the ferromagnet. This field spatially changes its sign corresponding to the typical scale of roughness. In the case of a trilayer the magnetization of one film can be frustrated by the laterally varying dipole field produced by the roughness of the second layer, and vice versa. This can yield a magnetic configuration where a perpendicular arrangement of the magnetization directions of the two films is energetically favored. As proposed by Demokritov *et al.*[20] one can assume a periodical roughness with period L and terrace height δ as illustrated in Fig. 4. From the exact form of the dipole field produced by this roughness the authors find that the energy of the magnetic-dipole coupling per unit area is proportional to $\cos^2\Theta$. The corresponding biquadratic coupling strength is

$$-J_2=\frac{M^4\delta^2L}{2\pi A'}\sum_{m=1}^{\infty}\frac{(-1)^{m-1}}{(2m-1)}\exp\left[-\frac{4\pi d}{L}(2m-1)\right]\times\left\{2-\exp\left[-\frac{8\pi D}{L}(2m-1)\right]\right\}, \tag{2}$$

where

$$A'=A+\frac{K_1L^2}{2\pi^2}$$

with the intralayer ferromagnetic stiffness A and the magnetic anisotropy constant K_1. We would like to emphasize that the coupling strength depends on the characteristic length L of the interface roughness and of course is independent of the spacer material. The leading term is an exponential in $-4\pi d/L$ and therefore in the case of $L\gg d$ the interaction is of comparatively long range.

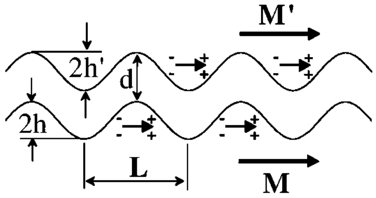

FIG. 5. Schematic illustration of Néel's ''orange peel'' effect (Ref. 31). Two magnetic films are separated by a nonmagnetic spacer of thickness d. Both interfaces with a sinusoidal roughness of period L and amplitude h are perfectly correlated.

Laterally varying dipolar fields created by interface roughness can also lead to a constructive interference and thus to a parallel or ''positive'' magnetostatic coupling as was earlier proposed by Néel.[31] The starting point is a rough substrate which exhibits a topography similar to an ''orange peel.'' In contrast to Demokritov *et al.*, Néel has assumed that the subsequently prepared nonmagnetic and magnetic layers adopt the topography of the substrate and consequently both interfaces are perfectly correlated, as is illustrated in Fig. 5. The magnetic dipoles of the two ferromagnetic layers are in perfect registry but shifted by one half

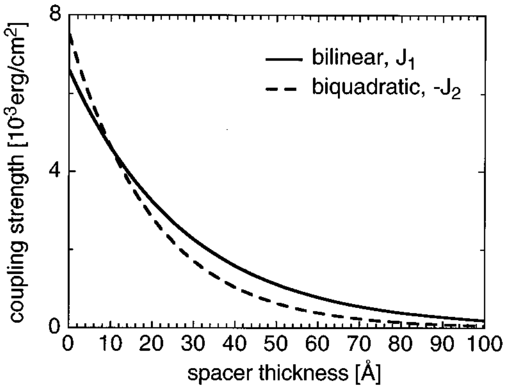

FIG. 6. Calculated dipolar coupling strengths for bilinear (solid line) and biquadratic (dashed line) coupling versus interlayer thickness as described in the text.

period. It therefore is plausible that magnetic flux closure or, in another picture, magnetostatic-repulsion and attraction between the dipoles results in a parallel alignment of the ferromagnetic films. Néel extends this one-dimensional model to the case where the roughness is periodical in two dimensions, which is more appropriate to describe the present experimental situation. He finds[31] that the correlated roughness at the two interfaces of the spacer layer leads to a contribution to the bilinear coupling with the following coupling strength:

$$J_1 = \frac{\pi^2}{\sqrt{2}L} hh'MM' \exp(-2\sqrt{2}\pi d/L). \qquad (3)$$

This bilinear coupling is quite similar to the biquadratic one described above. In particular the attenuation with increasing spacer thickness d is nearly the same for both effects.

Next we quantitatively evaluate the two roughness-induced dipolar coupling strengths J_1 and J_2 described above in Eqs. (3) and (2), respectively. We take the roughness parameters δ, h, and L from our structural analysis by STM. A decisive point for the quantitative interpretation of our observations is the correlation of roughness of the two interfaces on each side of the spacer layer. First we assume complete correlation of the two interfaces and compute the bilinear interaction [Eq. (3)], and then we assume that the two interfaces are entirely uncorrelated and calculate the corresponding biquadratic interaction [Eq. (2)]. Figure 6 shows the resulting bilinear (solid line) and biquadratic (dashed line) coupling strengths. As magnetic parameters we use known bulk Fe values of $A = 2\times10^{-6}$ erg/cm, $K_1 = 4.5\times10^5$ erg/cm^3, and $M = 1746$ μ/cm^3, and for the interfacial roughness we take the values of $2h = \delta = 5$ Å and $L = 20$ nm from our STM study. As illustrated in Fig. 6 both mechanisms exhibit about the same coupling strengths and nearly the same interlayer thickness dependences. Ferromagnetic coupling is preferred if the roughness at both interfaces is correlated and perpendicular alignment is preferred in the uncorrelated case. From these facts we draw the conclusion that the observed transition from ferromagnetic alignment at small spacer thicknesses to perpendicular alignment at higher thicknesses is caused by the loss of correlation of roughness at the two interfaces with increasing spacer thickness. For the particular roughness parameters found by STM, namely typical heights of 5 Å with typical widths of 200 Å, it is fair to assume that correlation does occur but only for thin spacer layers, where the adlayers are adopting the topography of the substrate. With increasing spacer thickness the correlation is likely to disappear. Therefore the bilinear coupling dominates over the biquadratic one for thin layers, while the loss of correlation for thicker interlayers results in preferred biquadratic coupling. The values of J_1 and J_2 of the order of a few 10^{-3} erg/cm^2, on the other hand, together with the absence of oscillatory coupling set an upper bound for the RKKY-type interaction of about 0.002 erg/cm^2 in this amorphous alloy.

In order to further illustrate the observed transition from the ferromagnetic to perpendicular alignment we as a first approximation assume a linear decrease of the correlation with increasing thickness. For spacers thicker than 60 Å the interfaces are believed to be uncorrelated. Qualitative estimates of the corresponding couplings are depicted in the bottom panel of Fig. 2. As expected, the bilinear term is damped (solid line) while the biquadratic one becomes more and more important (dashed line). For small spacers the coupling is strong and predominantly ferromagnetic. At a spacer thickness of about 30 Å the coupling changes from parallel to perpendicular alignment and the biquadratic coupling dominates over the bilinear one. The biquadratic coupling is quite small, even if the roughness is assumed to increase by a factor of 2 with increasing spacer thickness (dashed-dotted line). This qualitative thickness dependence well reflects the observed coupling strength, which for small couplings is approximately proportional to the spin polarization.

VI. SUMMARY

In this study we report that the coupling between two ferromagnets across an amorphous AuSn spacer is ferromagnetic at spacer thicknesses below 30 Å and favors 90° alignment above 50 Å. Between 30 and 50 Å the coupling continuously changes from ferromagnetic to 90° and remains in this perpendicular alignment for AuSn spacers up to least 100 Å. Following models by Demokritov *et al.*[20] and Néel[31] and using the roughness parameters directly obtained from STM studies, we have confirmed that the results can be understood in terms of roughness-induced magnetic dipolar interactions. Furthermore the absence of oscillatory exchange coupling across an amorphous spacer material supports the commonly accepted theories which strongly rely on the periodicity of the spacer. The quantitative analyses of the dipolar interactions yield an upper bound of the exchange interaction of 0.002 erg/cm^2 across this amorphous spacer.

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