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Long-range oscillatory exchange interaction between antiferromagnetic FeMn layers across a Cu spacer

J. W. Cai* and W. Y. Lai

State Key Laboratory of Magnetism, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China

J. Teng

Department of Materials Physics, University of Science and Technology Beijing, Beijing 100083, China

F. Shen and Z. Zhang

Beijing Laboratory of Electron Microscopy, Institute of Physics, Chinese Academy of Sciences, Beijing 100080, China

L. M. Mei

School of Physics and Microelectronics, Shandong University, Jinan 250100, China

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The exchange interaction between antiferromagnetic FeMn layers across a Cu spacer is studied by employing the exchange bias as a probe in multilayers of "NiFe/thin FeMn/Cu/thick FeMn." With variation of the Cu spacer's thickness, the indirect exchange interaction, monitored through the response of the exchange bias, oscillates with a period of approximately 18–20 Å, about twice that for ferromagnetic films separated by a Cu spacer. This result shows that long-range oscillatory exchange interaction is a basic and universal feature in both metallic ferromagnetic layers separated by nonmagnetic metals and metallic antiferromagnetic layers separated by a nonmagnetic metal, due to the quantum interferences induced by the spin-dependent interface reflection of Bloch waves with different oscillating periods originating from the difference in interface reflection conditions between ferromagnetic and antiferromagnetic spin ordering.

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The discovery of exchange coupling between Fe films separated by a thin Cr spacer layer[1] and its oscillatory behavior as the Cr thickness is varied[2] together with its relevance to giant magnetoresistance[3] have triggered a large number of experimental and theoretical investigations on multilayers consisting of different transition metal ferromagnets (FM's) and nonmagnetic (NM) spacers. By now it has become a well-understood general phenomenon that ferromagnetic layers of Fe, Co, Ni and their alloys separated by most any 3*d*, 4*d*, or 5*d* transition metal spacers[4–9] exhibit an exchange coupling that oscillates as a function of the spacer thickness with a period of approximately 10 Å (an exception, Cr). Antiferromagnetism, as the counterpart of ferromagnetism, originates from the same fundamental mechanism, i.e., the quantum-mechanical exchange interaction.[10] From a more general physical principle point of view, it is no doubt a critical question whether an exchange interaction can be propagated between metallic antiferromagnets (AF's) across a nonmagnetic metal spacer, just as in full-metal systems of the FM/NM/FM type.

While dealing with nanostructured AF's, people usually encounter the experimental difficulty of the insufficient sensitivity or resolution for most magnetometry and magnetic microscopy techniques. Fortunately, the unidirectional anisotropy of an FM layer adjacent to an AF layer, namely, exchange bias,[11] is readily measured and has been used to indirectly probe the properties of AF's, including the determination of the AF anisotropy,[12] spin flop field,[13] AF surface order parameter,[14] and AF domains.[15] We proposed that exchange bias might be employed to probe the interlayer exchange interaction between AF's in elaborate multilayers of "FM/AF(1)/NM/AF(2)." As is well known, exchange bias declines when the temperature approaches blocking temperature,[16,17] and it also evolves with the thickness of AF when the AF material is below a critical thickness,[18,19] both of which mean that exchange bias is sensitive to antiferromagnetism of the AF while the antiferromagnetism is diluted to some extent. Therefore, if long-range exchange interaction exists between AF's, the exchange bias in the structure of "FM/thin AF/NM/thick AF" is expected to vary in a certain way as the spacer thickness changes, considering that the dilute antiferromagnetism of the thin AF would be modified by the additional exchange interaction of AF's mediated via spacer.

The γ-FeMn alloy is a typical AF used for exchange bias, which has been extensively investigated since it was initially exploited as a domain stabilizer twenty years ago. So the knowledge about FeMn is rich and well understood. In this work, using "NiFe/thin FeMn/Cu/thick FeMn" multilayers, we have observed that the exchange bias oscillates with Cu thickness with a period of approximately 18–20 Å at a fixed FeMn thickness. This is the first experimental evidence of a long-range oscillatory exchange interaction between antiferromagnetic alloy layers across a metal spacer with a period approximately twice that of ferromagnetic films separated by a nonmagnetic spacer.

Films with a structure of "Ta buffer (40 Å)/NiFe (100 Å)/thin FeMn/Cu (8–48 Å)/thick FeMn/Ta (30 Å)" were grown in Ar at 0.5 Pa on water-cooled substrates of corning glass or native oxide-coated Si wafer in a

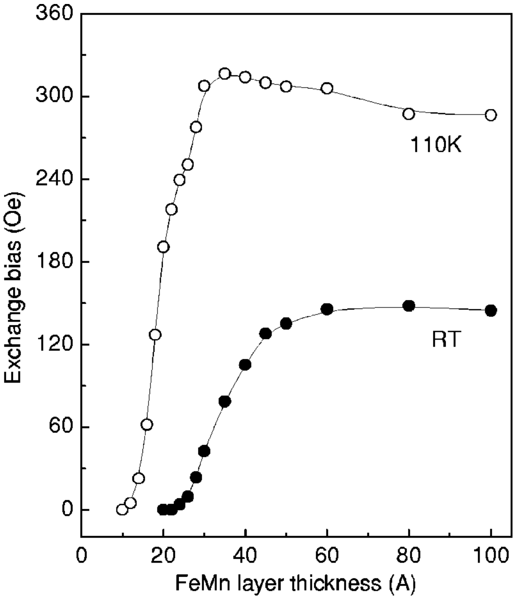

FIG. 1. Dependence of exchange bias on FeMn thickness at room temperature and 110 K for films Ta (40 Å)/NiFe (100 Å)/FeMn (10–100 Å)/Cu (60 Å)/Ta (30 Å).

multisource dc magnetron sputtering system. A static field of about 300 Oe was applied to the film plane during deposition to produce the exchange bias. The base pressure of the vacuum system is better than 3×10^{-5} Pa. Targets of $\mathrm{Ni}_{80}\mathrm{Fe}_{20}$, $\mathrm{Fe}_{50}\mathrm{Mn}_{50}$ alloys and Cu and Ta (purity 99.9%) were used to grow NiFe, FeMn, Cu, and Ta films, respectively, and the sputtering rates were of 1.0–1.2 Å/sec. Sets of up to eighteen samples were prepared at a time. M-H curves were measured by using a DMS Model 4 HF vibrating sample magnetometer (VSM) from ADE technologies with a field resolution of 0.01 Oe. The crystalline structure of the films was checked in a Rigaku x-ray diffractometer using Cu $K\alpha$ radiation, and the detailed microstructures of the films were examined in a Tecnai F20 transmission electron microscope (TEM) equipped with a Gatan imaging filter (GIF) with spatial resolution of 10 Å. The magnetoresistance of the films was studied by using the dc four-point probe method.

As stated above, the appropriate dilute antiferromagnetism of a thin FeMn layer, which is governed by its thickness as well as its temperature, is crucial to the present study. So a series of films Ta (40 Å)/NiFe (100 Å)/FeMn (10–100 Å)/Cu (60 Å)/Ta (30 Å), was first prepared to determine a thickness range for the thin FeMn layer. Figure 1 shows the dependence of exchange bias on FeMn thickness at room temperature and at low temperature (110 K). These results are consistent with the published data.[18,19] Note that exchange bias at room temperature appears at an FeMn thickness of 24 Å, and quickly increases with the increase of FeMn thickness up to 45 Å, finally reaching its saturated value at some 60 Å, whereas at 110 K the onset of the exchange bias occurs at 12 Å and peaks at 35 Å before falling to a constant value. It is appropriate to define the dilute AF limit as the exchange bias reducing to a value below a maximum of 60%, then an FeMn layer could be take as a dilute AF when its thickness is below 35 Å at room temperature, but the thickness must be smaller than 20 Å at 110 K. On the other hand, the FeMn layer with thickness beyond 80 Å is certainly a strong AF, both at room temperature and low temperature. Based upon these results, thirteen sets of films with thin FeMn at every 2 Å increment from 12 to 36 Å and thick FeMn of 150 Å were fabricated for a full investigation.

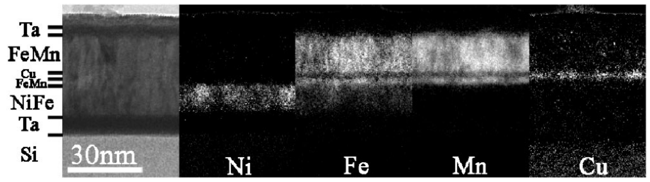

FIG. 2. Bright field image of a cross section of the multilayer Ta (40 Å)/NiFe (100 Å)/FeMn (26 Å)/Cu (19 Å)/FeMn (150 Å)/Ta (30 Å) and the corresponding elemental maps of Ni, Fe, Mn, and Cu by using their $L_{2,3}$ edges, respectively.

The film structure characterization was performed. X-ray diffraction (XRD) on samples shows that the constituent layers of NiFe, FeMn, and Cu were strongly (111) textured and coherently grown because of the small lattice mismatches (~2%) with all layers having a face-centered-cubic structure. The columnar growth of the films was further verified by cross-section TEM observations as shown in the left panel of Fig. 2. However, one could hardly identify the multilayer structure from the bright field image of TEM due to the close atomic numbers and the consequent small difference in electron scattering ability. Elemental mapping based on electron energy-loss spectroscopy (EELS) and the three-widow method[20] with spatial resolution of 10 Å was thus used to solve the composition-related problem. Corresponding to the bright field image of a cross-section view of a typical multilayer Ta (40 Å)/NiFe (100 Å)/FeMn (26 Å)/Cu (19 Å)/FeMn (150 Å)/Ta (30 Å), elemental mappings of Ni, Fe, Mn, and Cu by using their EELS $L_{2,3}$ edges are shown in Fig. 2 together. It should be noted that the Fe concentration is only 20% in the NiFe layer and 50% in the FeMn layers, therefore the contrast is not uniform in Fe maps of NiFe and FeMn layers. Compared with the bright field image, elemental mappings of Ni, Fe, Mn, and Cu clearly show the multilayer structure of the film, indicative of an ideal layer structure of two AF layers separated by a thinner Cu layer of 19 Å.

The room-temperature magnetic measurements show that appreciable exchange bias appears for samples with thin FeMn above 16 Å at a Cu thickness of 8 Å, and it decreases monotonically to zero with the increase of Cu thickness up to at most 19 Å when thin FeMn is below 24 Å. This indicates that the exchange bias of a dilute FeMn layer is enhanced by a thick FeMn with a thin Cu layer sandwiched in between. Moreover, a rather astonishing phenomenon emerges when thin FeMn is just beyond the onset of exchange bias of a single FeMn layer. Figure 3 shows the dependence of exchange bias on the thickness of the Cu spacer for two sets of samples Ta (40 Å)/NiFe (100 Å)/FeMn (26 or 28 Å)/Cu (8–48 Å)/FeMn (150 Å)/Ta (30 Å). It is noted that the exchange bias is fairly high when Cu is thin, and most interesting, a broad bump comes out after the rapid decrease of

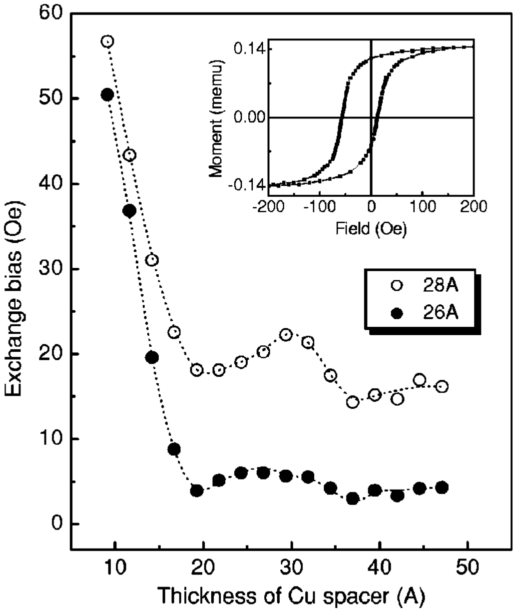

FIG. 3. Dependence of exchange bias on the thickness of Cu spacer at room temperature for two sets of samples Ta (40 Å)/NiFe (100 Å)/FeMn (26 or 28 Å)/Cu (8–48 Å)/FeMn (150 Å)/Ta (30 Å). The inset depicts a representative hysteresis loop for the multilayer of Ta (40 Å)/NiFe (100 Å)/FeMn (28 Å)/Cu (29.4 Å)/FeMn (150 Å)/Ta (30 Å).

the exchange bias for each set of samples, and the exchange bias shows an obvious oscillation with the thickness of Cu spacer at a definite period of about 18–20 Å, which is almost twice that for exchange coupling between ferromagnetic layers separated by a Cu spacer.[2] The above results indicate that long-range exchange interaction with a distinctive feature indeed exists between AF's across a nonmagnetic spacer. We would like to point out that, except for a different amount of loop shift, the hysteresis loop Ta/NiFe/thin FeMn/Cu/thick FeMn/Ta mutilayers are similar to those of Ta/NiFe/thin FeMn/Cu/Ta films. The inset of Fig. 3 depicts a representative hysteresis loop corresponding to the curve peak for the set of samples Ta (40 Å)/NiFe (100 Å)/ FeMn (28 Å) / Cu (8–48 Å) / FeMn (150 Å) / Ta (30 Å). One can note that the hysteresis loop is symmetrical on magnetization reversal from $+M$ to $-M$ and vice versa.

Since the exchange bias for the present structure is primarily determined by the thin FeMn, and the response of the exchange bias to the interlayer interaction of AF's is a secondary effect, the exchange bias is susceptible to the extra long-range exchange interaction only for dilute FeMn at a narrow thickness range just above the onset of exchange bias of a single FeMn layer. If the thin FeMn layer is rather thin, the weak interlayer exchange interaction at large Cu thickness cannot reinforce the much diluted antiferromagnetism beyond the threshold of the exchange bias, and therefore, no oscillatory exchange bias was observed for rather thin FeMn layers as mentioned earlier. On the other hand, if the thin FeMn layer becomes thick enough to establish a sufficient exchange bias by itself, the long-range exchange interaction cannot alter the exchange bias any more. In fact, a further increase in the thickness of the thin FeMn leads to a less clear oscillatory variation of the exchange bias, and no oscillating behavior is observable when thin FeMn beyond 34 Å, which almost coincides with the dilute FeMn limit that we defined for room temperature. Parenthetically, the fabrications and tests of all samples were repeated at least three times to ensure the reliability of the experimental data, and similar results were obtained across the board.

Even so, one may argue that the observed oscillatory exchange bias is caused or at least partially comes from the interaction between the thick AF and FM through the thin AF+NM layer. Indeed oscillatory exchange bias was reported in NiFe/Cu/NiO,[21] and the CoO was found to be coupling to NiFe through metallic spacers at relatively long range.[22,23] However, insulating AF's belong to a completely different type of system from metallic AF's in view of their electronic structures. It is improper to apply observations of insulating AF's to the present metallic system. In fact, all-metal FM/NM/AF systems[24,25] showed an interlayer exchange interaction within only a few Å, and the interlayer exchange coupling disappears for NiFe/Cu/FeMn system[25] when the Cu layer is thicker than 6 Å. Therefore, it is definitely impossible for the NiFe layer to be exchange biased by the thick FeMn layer across a huge spacer (26 or 28 Å FeMn plus 19–45 Å Cu) in the multilayers of NiFe/thin FeMn/Cu/thick FeMn. One may worry about another effect: a change of the microstructure of the thin FeMn layer due to changes in growth conditions correlated with Cu thickness, which could play a role in the observed oscillatory exchange bias. We would like to stress that although varying the thickness of the Cu buffer could lead to significant differences in the exchange bias properties of NiFe/FeMn bilayers,[26] the intermediate NM Cu layer is grown above the thin FeMn layer in the present case, so the effect of the Cu spacer on the microstructure of the thin FeMn layer should be very little. We actually examined a series of multilayers with the thick FeMn removed, i.e., Ta (40 Å)/NiFe (100 Å)/FeMn (28 Å)/Cu (8–48 Å)/Ta (30 Å), and there is little difference in the exchange bias properties among these samples. Therefore, the oscillatory exchange bias in the multilayers NiFe/thin FeMn/Cu/thick FeMn is most likely from the exchange interaction between the thin and thick FeMn layers.

The exchange coupling in ferromagnetic multilayers becomes stronger as temperature decreases.[27,28] The oscillatory behavior of the exchange bias at 110 K appears for the set of samples with thin FeMn of 14 Å as shown in Fig. 4. However, the samples with thin FeMn of 26 or 28 Å had the exchange bias on the order of 300 Oe, with only very slight changes for all thicknesses of the Cu layer after cooled down to 110 K. Since the dilute FeMn limit at 110 K is 20 Å, it is not strange that the samples with thin FeMn of 26 or 28 Å do not exhibit oscillatory exchange bias with varying Cu thickness, which could also be understood from Fig. 1, where a layer of FeMn at 26 or 28 Å is somehow nearby the peak at 110 K. We would like to point out that a cooling field of any intensity from 1 to 10 kOe makes no difference in the exchange bias field for the samples, and the low-temperature results shown in this paper were obtained under the same cooling process with the cooling field of 1 kOe. Carefully checking the variation of exchange bias at 110 K for the samples with the thin FeMn of 14 Å, one can notice that the

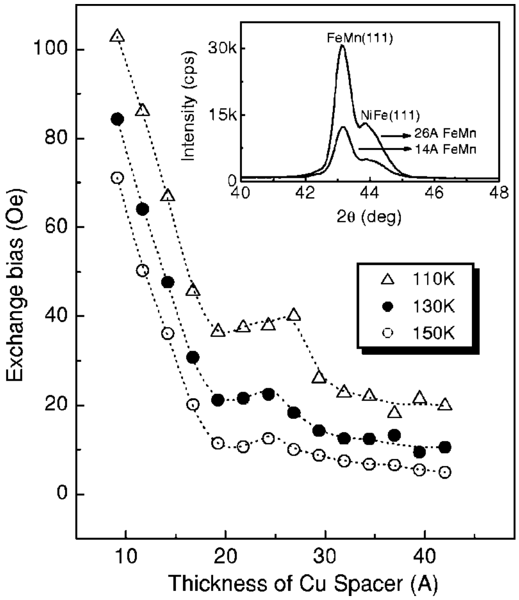

FIG. 4. Dependence of exchange bias on the thickness of Cu spacer at low temperature for samples Ta (40 Å)/NiFe (100 Å)/FeMn (14 Å)/Cu (8–48 Å)/FeMn (150 Å)/Ta (30 Å). The inset depicts XRD spectra for two samples Ta (40 Å)/NiFe (100 Å)/FeMn (14 and 26 Å)/Cu (19 Å)/FeMn (150 Å)/Ta (30 Å).

oscillating period is not so evident and slightly less consistent, which seems to result from degraded structure. The inset of Fig. 4 is XRD patterns for samples with 19 Å Cu and thin FeMn of 14 and 26 Å, respectively. It is very clear that the peaks of both FeMn(111) and NiFe(111) decrease to less than half of their values when the thin FeMn is changed from 26 to 14 Å. We would like to point out that the oscillatory variation of the exchange bias gradually disappears with an increase of temperature, which may come partly from the reduction of the long-range exchange interaction along with the further weakening of the antiferromagnetism of thin FeMn itself due to thermal fluctuation.

In an FM/NM/FM system, the interlayer exchange interaction leads to the spins of the FM layers in either the parallel or antiparallel state, depending on the thickness and specific electronic structure of the NM layer. We extended the well-established theoretical models[7,8] aimed at this general phenomenon in the FM/NM/FM system to interpret the interlayer exchange interaction of AF's in the trilayer of AF/NM/AF. It should be emphasized that there are also two different spin alignment configurations at the two interfaces of AF/NM/AF, i.e., ↑↓/NM/↑↓ vs ↑↓/NM/↓↑, where the arrows nearest and next nearest to the slash represent the directions of the interface spin, and the neighboring interface spin, respectively. Because of the spin-dependent reflections of Bloch waves at the two interfaces, the quantum interference states confined in the spacer are different for these two spin alignment configurations, and the corresponding energies, which oscillate with spacer thickness, are also different. The difference of these oscillatory energies makes the oscillatory interlayer exchange interaction. Since exchange bias primarily depends on the interfacial AF spin structure and AF domains in FM/AF bilayers, the oscillatory interlayer interaction of AFs can be converted to the exchange bias effect in FM/AF(1)/NM/AF(2) only when the AF(1) is in the dilute AF range with its domains or spin structure susceptible to the interlayer interaction. It should be pointed out that, for most experimental investigations, the detailed information about the AF spin structure and AF domains is still lacking and difficult to achieve even for FM/AF bilayers. Alternatively, the Neél temperature varies with thickness for a dilute AF because of the finite size effect.[29] An understandable picture for the oscillatory exchange bias effect observed in FM/dilute AF/NM/strong AF is that the effective Neél temperature or effective thickness of the dilute AF is modified by the long-range oscillatory exchange interaction. However, the interlayer exchange interaction in AF/NM/AF might be much weaker than that in FM/NM/FM, and one should not expect a significant effect of the interlayer exchange interaction on the effective Neél temperature of the dilute AF. From the spin alignment configurations at the interface of AF/NM/AF, ↑↓/NM/↑↓ and ↑↓/NM/↓↑, the exchange interactions are partially cancelled for the interface spin and the neighboring interface spin, in contrast with that for FM/NM/FM.

As for the oscillatory period for the AF/NM/AF system, the unit cells of AF and NM lattices must be doubled in x, y, and z directions to ensure the existing models[7,8] directly applicable with in-plane translational invariance remaining unbroken by the alternative arrangement of the antiparallel moments in the AF lattice. Since the period is set by the extremal spanning vector of the Fermi surface of the spacer layer material, which is now shortened with the shrinking of the first Brillouin zone because of the doubled lattice, the period of the oscillatory exchange interaction between AF's turns out to be 21.3 Å through a simple estimation, in good agreement with the experimental findings. The strength of the coupling depends both on the geometry of the Fermi surface and on the reflection amplitudes for electrons scattering from the interfaces between AF and NM. A determination of the coupling strength is limited by the nature of the exchange bias, and needs further exploration.

Finally, it is natural to think of magnetoresistance if one observes oscillatory exchange coupling in magnetic multilayers. However, only anisotropic magnetoresistance (AMR) of the permalloy layer is evident (MR below 1%) for "NiFe/thin FeMn/Cu/thick FeMn" multilayers, which means that the magnetoresistance effect related to the oscillatory exchange interaction between the FeMn layers should be extremely small or nonexistent.

In conclusion, we have found convincing experimental evidence of oscillatory exchange interaction between antiferromagnetic FeMn layers across a Cu spacer with a period approximately twice that of ferromagnetic multilayers. This result shows that long-range oscillatory exchange interaction is a basic and universal feature in metallic FM/NM/FM and AF/NM/AF due to the quantum interferences induced by the spin-dependent interface reflection of Bloch waves with different oscillating periods originating from the different interface reflection conditions for ferromagnetic and antiferromagnetic spin ordering.

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*Electronic address: jwcai@aphy.iphy.ac.cn

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