ORDER, DISORDER, AND PHASE TRANSITION =

Effect of the Nonmagnetic Layer in a Co/Cu/CoO Trilayer Structure on the Exchange Coupling in It

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Abstract—The dependence of the exchange bias of epitaxial single-crystal Co/Cu/CoO trilayer films on the copper layer thickness and temperature is studied. The exchange bias of the hysteresis loops of the ferromagnetic cobalt layer as a function of the copper layer thickness is found to have a well-pronounced oscillating character. The oscillations manifest themselves over the entire temperature range in which an exchange bias takes place (77–220 K). The complex variation of the oscillation amplitude with the nonmagnetic layer thickness can be explained by the superposition of two interlayer exchange coupling oscillation periods ($\lambda_1 \approx 10-11$ Å, $\lambda_2 \approx 20$ Å) having different amplitudes and temperature dependences.

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INTRODUCTION

The phenomenon of the unidirectional anisotropy induced by the exchange coupling at the interface between two materials with different types of magnetic ordering, namely, ferromagnetic (F) and antiferromagnetic (AF) materials, has attracted the attention of researchers from a standpoint of both the fundamental problems of coupling in heterogeneous systems and possible practical applications. Although this phenomenon has long been studied [1], there is no generally accepted concept of the nature of this phenomenon. For example, there is no agreement among researchers regarding the type of this coupling, i.e., short-range [2] or long-range [3] coupling. The assumptions that a unidirectional anisotropy results from the exchange coupling between the neighboring spins of F and AF materials at the interface point to the possibility of such coupling in the case of F and AF materials separated by a conducting layer of a nonmagnetic (NM) material, as in the case of F/NM/F structures. Indeed, the long-range exchange coupling through the conduction electrons of a nonmagnetic layer between F layers in magnetic multilayer structures $(F/NM)_n$ and related structures has been much studied [4–6]. This coupling has an oscillating character as a function of the NM layer thickness and is well explained by the theories based on an indirect exchange (RKKY coupling) model [7, 8].

On the other hand, the coupling between F and AF layers is much more complex: the exchange bias field was found to strongly depend on a spin structure at the interface. The interface roughness, the presence of domain walls, and the relative orientation of the spins of F and AF layers significantly affect the exchange coupling and make it difficult to compare experimental and calculated results [9, 10].

The first experimental work dealing with the interaction of F and AF layers through an NM layer revealed a long-range coupling between them [3]. However, this coupling had no oscillating character, as would be expected for an RKKY-type coupling, and the weakening of the interlayer coupling had an exponential character. In later works, researchers revealed a nonmonotonic dependence of the exchange field on the NM layer thickness, which was attributed to the interface roughness [2], and an oscillating dependence against the background of a strong monotonic weakening of the exchange bias at a period of about one monolayer [11]. The authors of [12] detected a pronounced oscillating dependence with a period of about 11 Å (copper was an NM material) at temperatures close to the critical temperature of the AF layer, which agrees well with theoretical predictions for such an RKKY coupling. However, at lower temperature, the oscillating dependence is superimposed on an exponential decrease in the exchange bias, whose nature is still unknown. This exponential contribution has a strong temperature dependence. The authors of [12] believe that such a temperature-dependent contribution can be caused by the presence of bridges between F and AF layers (in pores of a polycrystalline interlayer), which result in a direct exchange coupling. However, oscillations were not detected in experiments performed on a Co/Au/CoO structure [13]. The authors of [14] also detected an oscillating dependence of the exchange bias field on the copper layer thickness in Fe/Cu/CoO films, which were grown by molecular-beam epitaxy and the antiferromagnetic layer of which consisted of misoriented (presumably) amorphous nanoparticles. In [15], the coupling between two AF FeMn layers separated by a copper layer was studied, and an exchange coupling oscillation period of 18–20 Å was detected. In [16], we investigated polycrystalline NiFe/Cu/IrMn samples and found exchange bias oscillations with a period of 8 Å and a decrease in the exchange bias field against the background of an exponential decrease. Nevertheless, some researchers [2, 17] deny a long-range character of the exchange coupling and present experimental results indicating a short-range character of this phenomenon.

EXPERIMENTAL

We studied a series of single-crystal Co/Cu/CoO samples formed by magnetron sputtering onto an oriented MgO(001) substrate. The sputtering conditions (the substrate temperature, the component sputtering rate) were chosen to ensure epitaxial growth of a film structure on an MgO(001) substrate. Sputtering was carried out in an Ar atmosphere at a working pressure of 9×10^{-4} Torr, and the base vacuum was 5×10^{-6} Torr. We grew Co(150 Å)/Cu(d_{Cu})/CoO(150 Å)/MgO structures in which the copper layer thickness was varied in the range $d_{\rm Cu} = 2.5 - 40$ Å. The grown structures satisfied the epitaxial orientation relationships fcc $MgO(001)[100] \parallel fcc CoO(001)[100]$ || fcc $Cu(001)[100] \parallel fcc Co(001)[100]$. The layer thicknesses were specified by the time and rate of sputtering of the corresponding materials and were controlled by X-ray fluorescence. The accuracy of measuring the layer thickness was better than one atomic layer for each component.



Fig. 1. X-ray diffraction pattern of a Co/Cu(25 Å)/CoO/MgO sample. (inset) The (002) reflection of fcc cobalt.

RESULTS AND DISCUSSION

Figure 1 shows the typical X-ray diffraction pattern of the samples (table lists the lattice parameters of bulk Co, Cu, CoO, and MgO samples). The X-ray diffraction pattern has the (002) reflection of fcc cobalt, whose position corresponds to the reflection of bulk fcc cobalt, and the (002) and (004) reflections of single-crystal MgO substrate. The absence of reflections from atomic planes of the cobalt oxide CoO layer is caused by the closeness between the lattice parameters of CoO and MgO (4.26 and 4.213 Å, respectively). These reflections are overlapped with high-intensity reflections of MgO, and the absence of reflections from atomic planes of copper is caused by a small copper layer thickness. In experiments where a CoO layer had a significantly larger thickness, the (002) and (004) reflections of CoO with a lattice parameter of 4.26 Å were present. The broadening of the (002) reflection of fcc cobalt can be explained by the size effects induced by a small cobalt layer thickness. However, the calculation by the Scherrer formula [19] with allowance for the real full width at half-maximum ($\beta =$ 2°) yields a cobalt layer thickness $d_{\rm Co} = 45$ Å, which is approximately three times smaller than the value determined from the technological sputtering parameters and the X-ray fluorescence data. This line broad-

Lattice parameters a (distance between (001) planes) for the system components taken from data base in [18]

Component	MgO	CoO	Cu	Co
<i>a</i> , Å	4.213	4.26	3.607	3.554



Fig. 2. Hysteresis loops of Co/Cu/CoO/MgO films recorded at various copper layer thicknesses.

ening is most likely to be caused by deformationinduced lattice distortions. Specifically, the large misfit between the lattice parameters of copper (and cobalt with a similar lattice parameter) and CoO oxide (3.607 Å (3.554 Å) and 4.26 Å, respectively) in the case of epitaxial growth results in significant stresses at the CoO/Cu interface. Because of a thin copper layer, these stresses can be retained in the copper layer and, in turn, cause tensile stresses of the cobalt lattice at the Co/Cu interface, which relax in the bulk of the cobalt layer. The calculation demonstrates that the average change in the lattice parameter of cobalt that can cause such a change in the line width is $\Delta d = 0.03$ Å;



Fig. 3. Exchange bias of a hysteresis loop vs. the copper layer thickness at various measurement temperatures.

i.e., we have $d_{\text{Co}(200)} = (1.77 \pm 0.03)$ Å (see [19]). However, it is impossible to perform angular analysis of the broadening distribution of diffraction lines in order to reveal the nature of broadening because of a preferred orientation of our samples and the absence of reflections from other atomic planes of cobalt (see [19]). Moreover, this reflection of cobalt can also be affected by the (002) reflection of copper, which causes an asymmetric profile of this diffraction line.

From a magnetic standpoint, the cobalt lavers are ferromagnetic with a pronounced biaxial magnetic anisotropy in the film plane, an anisotropy constant $K_1 \sim 3 \times 10^5$ erg/cm³, and easy axes parallel to [110] and $[1\overline{1}0]$ directions (as is determined by the magnetocrystalline anisotropy of fcc cobalt). To measure the exchange bias, the samples were magnetized at room temperature in a field of 3 kOe applied along one of the easy axes of the cobalt layer. They were then cooled in this field to the measurement temperature, and the temperature was fixed for a hysteresis loop to be measured. All hysteresis loops obtained under these conditions are rectangular and characterized by a bias toward negative (in the experimental geometry) fields, which is caused by the long-range exchange coupling of Co and CoO layers through a copper layer (see Fig. 2). The dependences of exchange bias H_E of a hysteresis loop on the copper layer thickness obtained at various temperatures are shown in Fig. 3. An oscillating character of the measured exchange bias is clearly visible. At low temperatures (below 200 K), at least three maxima in the $H_E(d_{Cu})$ curves at 10, 20, and 35 Å are well pronounced. A nonmonotonic character of



Fig. 4. Temperature dependences of the exchange bias for films with various copper layer thicknesses.

the change in the exchange bias (oscillation amplitude) as a function of the copper layer thickness should be noted; this character was not detected in the experimental dependences of the exchange bias obtained for both FM/NM/FM [6, 7] and FM/NM/AFM structures [12, 14]. This picture can be explained if we assume the presence of two exchange coupling oscillation periods, as in the case of single-crystal FM/NM/FM structures. Indeed, the existing theories of exchange coupling in FM/NM/AFM structures, which originated from works [7, 8], relate the exchange coupling oscillation period to the topology of the Fermi surface of the NM layer metal. For a (001) copper layer, these theories predict the coexistence of two oscillation periods, namely, short ($\lambda_1 =$ 4.7 Å) and long ($\lambda_2 = 10.6$ Å) periods, having different amplitudes. The experimentally measured values are $\lambda_1 = 4.5 \text{ Å and } \lambda_2 = 10 - 15 \text{ Å } [20, 21]$. Since our experiments were also performed on samples with a (001) copper layer used as a conducting interlayer, we can also assume the presence of two oscillation periods in our case. Moreover, significant stresses at the CoO/Cu interface, which induce lattice distortions, are inevitable in our case, where a copper layer grows epitaxially on the surface of the CoO layer, because of a significant misfit between the lattice parameters of the layers. Such a misfit either leads to uniform stresses in the laver or is removed through interfacial dislocations. Both mechanisms cause distortions in the crystal structure of the copper layer and, hence, distortions in the topological shape of the Fermi surface of the material. At least for the case of an FM/NM/FM system, this shape determines the detected exchange coupling oscillation periods.

Thickness dependences $H_E(d_{Cu})$ can be explained by the presence of two exchange coupling oscillation periods, namely, $\lambda_1 \approx 10-11$ Å and $\lambda_2 \approx 20$ Å, which have different amplitudes and temperature dependences. Figure 4 shows the exchange biases of films having various copper layer thicknesses. The decrease in the exchange bias for a film with $d_{Cu} = 20$ Å is seen to be much faster than in the other samples.

CONCLUSIONS

We studied single-crystal epitaxial exchange-coupled Co/Cu/CoO/MgO(001) structures and detected a nonmonotonic oscillating character of the exchange coupling in them, which can be explained by the presence of two oscillation periods having different amplitudes and temperature dependences. The exchange bias in such structures was shown to be controlled by introducing a conducting nonmagnetic layer between FM and AFM layers, which is important for designing modern magnetoelectronic devices for spintronics.

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