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Observation of nonmonotonic oscillatory interlayer exchange coupling in Co/Cu/CoO films with varying Cu spacer thickness

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We report our experimental observation of interlayer exchange coupling phenomena in CoO/Cu/Co trilayers with systematic variation of Cu spacer layer thickness as well as temperature. It has been found that there exists a clear indication of nonmonotonically varying oscillatory interlayer exchange coupling. The amplitude of oscillation increases, reaches to the maximum, and decreases with increasing Cu spacer thickness from 1 to 16 atomic layers for all temperature ranges between 70 and 200 K. © *2008 American Institute of Physics*. [DOI: 10.1063/1.2831390]

Exchange bias after the first discovery in 1950s (Ref. 1) has been observed in various kinds of systems having interfaces between ferromagnet and antiferromagnet.[2] In addition to the interest in exciting physics related to this phenomenon, it has recently attracted huge interest renewed mainly by its application to magnetic recording and sensor technology.[3–5] Interestingly, interlayer exchange coupling phenomenon has been reported to be oscillatory in ferromagnetic (FM) or antiferromagnetic (AFM) layers spaced by a nonmagnetic layer.[6–10] The oscillation has been found to depend on the temperature of the system[11] and the thickness of spacer layer.[6–9] More studies have been focused on two FM layer systems with nonmagnetic (NM) spacer layer such as in Co/Au/Co,[6] Co/Cu/Co,[7] and Fe/Cr/Fe,[12] where it has been reported that long-range exchange coupling decreases across the spacer. On the other hand, the first observation of oscillatory interlayer exchange coupling behavior in AFM/NM/FM trilayers with thickness variation of NM spacer has been reported in FeMn/Cu/FeNi,[13] later in NiO/Cu/NiFe,[14] and CoO/Cu/Co,[15] where a thermal contribution is considered to explain the oscillatory behavior in AFM/NM/FM system together with Ruderman–Kittel–Kasuya–Yosida—like coupling and interlayer dipolar interaction. However, very little has been known for the exact mechanism of oscillatory interlayer coupling in AFM/NM/FM system. For example, the existence of oscillatory behavior in AFM/NM/FM system is still controversial.[16,17]

In most systems having interlayer exchange coupling, oscillation amplitude seems to be decreased monotonically through the spacer with increasing spacer layer thickness. The decreasing tendency has been reported to be dependent on the spacer layer thickness t with t^{-1}, t^{-2}, or exponential decay.[18] The decreasing oscillation amplitude is mainly explained based on the geometry of the spacer Fermi surface and the extended states within the spacer layer.[18,19] However, in AFM/NM/FM system, the dependency of oscillating amplitude with variation of spacer layer thickness and temperature has not yet clearly understood. In this work, we report our systematic experimental investigation of interlayer exchange coupling behavior in CoO/Cu/Co (AFM/NM/FM) trilayer system with control of spacer layer thickness and measurement temperature. Clear indication of oscillatory interlayer exchange coupling is observed with a nonmonotonic variation of oscillation amplitude.

The CoO/Cu/Co trilayer has been prepared by magnetron sputtering with a base pressure of 5×10^{-6} Torr and Ar sputtering pressure of 9 mTorr. The bottom CoO layer with a thickness of 100 Å was deposited on MgO (111) substrate. Cu layer was grown on CoO as a spacer layer with thickness variation from 2.5 to 40 Å, which corresponds to 1–16 atomic layers, respectively. A 150 Å Co was deposited on top of Cu layer. External magnetic field was applied along a certain direction on the sample plane to induce an easy axis of Co layers during the deposition. X-ray diffraction (XRD) measurement on Co/Cu/CoO/MgO demonstrated epitaxial layer of single crystal fcc-Co (200) due to a pseudomorphic growth of Co on a single crystal MgO. Structural properties of the samples were studied by transmission electron microscopy. Magnetic hysteresis loops were measured by superconducting quantum interferometer device and vibrating sample magnetometer. Each sample was initially field cooled under the field of 5 kOe from the room temperature (~300 K) down to 70 K. The field-cooling process is expected to be working since the Néel temperature of CoO (T_N=290 K) is known to be lower than the room temperature. The field direction of the field cooling was chosen to be parallel to the induced easy axis of Co layer by the external field applied during the sample deposition. Exchange-biased hysteresis loops were measured for samples with various Cu spacer layer thicknesses at various temperatures.

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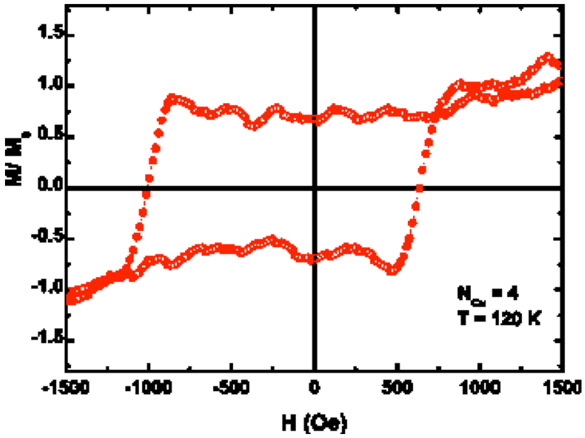

FIG. 1. (Color online) Typical magnetic hysteresis loop measured on the sample plane by vibrating sample magnetometer for the sample having four-layer thick Cu spacer (N_{Cu}=4) at the temperature of 120 K.

A typical example of an exchange-biased magnetic hysteresis curve is illustrated in Fig. 1. The curve in the figure is measured at 120 K for the sample having four layers of Cu spacer, exhibiting a clear sign of negative exchange shift. The exchange bias field as well as left and right coercivities (H_{ex}, H_{CL}, and H_{CR}, respectively) are determined from hysteresis curves and plotted in Fig. 2. In the figure, four representative cases are plotted for the sample with 2, 6, 8, and 12 Cu spacer layers. All other cases have similar trend, as shown in Fig. 2. The overall decrease of exchange bias is observed for all samples, as demonstrated in Fig. 2(c). The sample with two layers of Cu spacer exhibits a transitional behavior from negative to positive exchange bias with increasing the temperature, while all other thicker samples keep the negative exchange bias until the exchange shift vanishes around the blocking temperature. The origin of an exceptional oscillatory behavior of the sample with two layers of Cu spacer is unclear.[20] One possibility is that a discontinuous or disordered two layer thin Cu spacer film might cause the exceptional behavior. However, considering the clear XRD peak at fcc Co (200) position, the trilayers with a Cu spacer thicker than two layers are expected to have continuous Cu spacer layers on which 150 Å Co film is deposited.

The exchange biases for samples of different Cu spacer layer thicknesses are measured at various temperatures, as shown in Fig. 3. It is clear that there exists an oscillatory behavior for all measurement temperatures below 200 K. The period of the oscillation seems to be about four Cu layers (~10 Å) for all temperatures, which provides a clear experimental evidence for the existence of oscillatory behavior in the AFM/NM/FM trilayer system. The controversial experimental results reported so far for the existence of oscillatory interlayer exchange coupling in AFM/NM/FM trilayer[13,16,17] are theoretically not yet fully understood. In the present work, the H_{ex} exhibits no clear indication of oscillatory behavior depending on the temperature as in Fig. 2(c), and the oscillatory behavior exists only for the variation of Cu spacer layer thickness.

Moreover, the period of the oscillation (~10 Å) seems to be irrespective of the measurement temperatures within the measurement error, as demonstrated in Fig. 3. This implies that the origin of the oscillatory interlayer exchange behavior observed in the present study is not the thermal process proposed in case of NiO/Cu/NiFe.[14] The oscillation

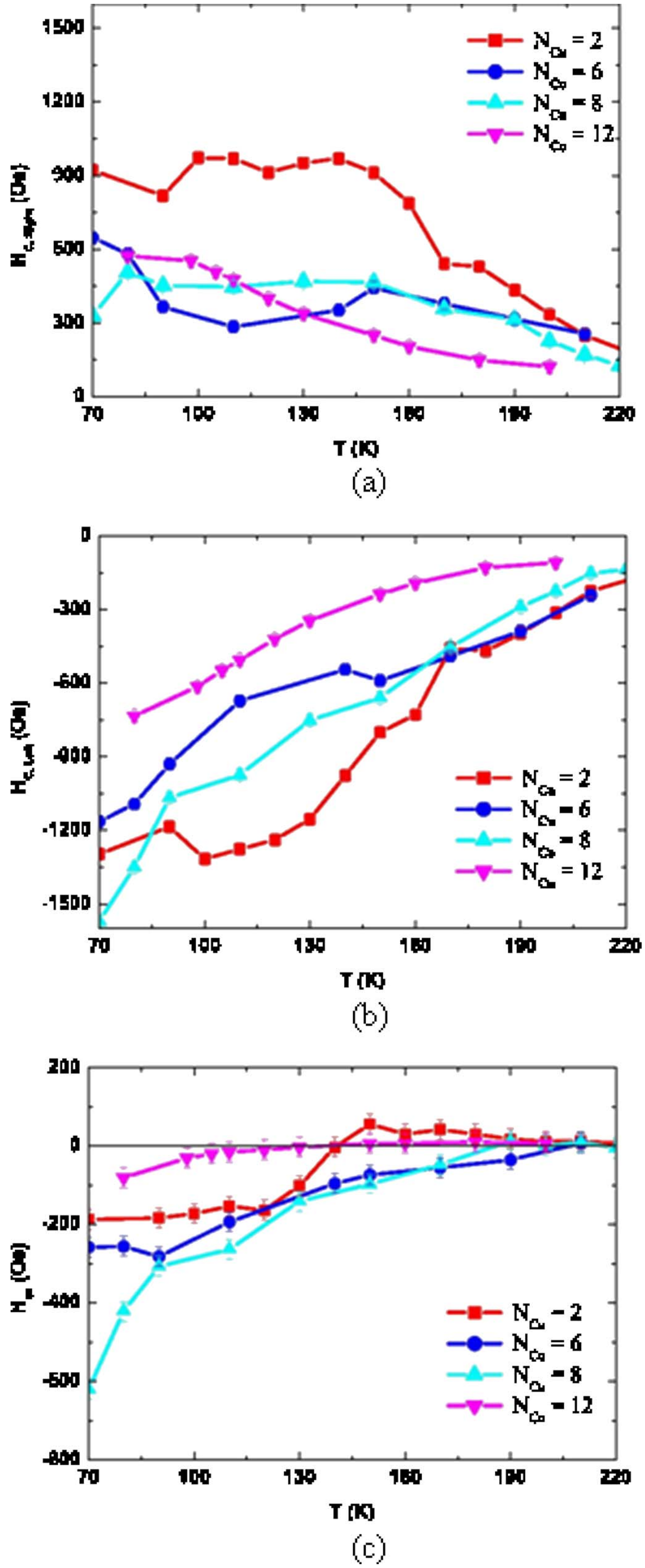

FIG. 2. (Color online) Typical example of (a) left ($H_{C,left}$), (b) right ($H_{C,right}$) coercivities, and (c) exchange bias (H_{ex}) depending on the temperature for the sample having 2, 6, 8, and 12-layer thick Cu spacer (N_{Cu}=2, 6, 8, and 12).

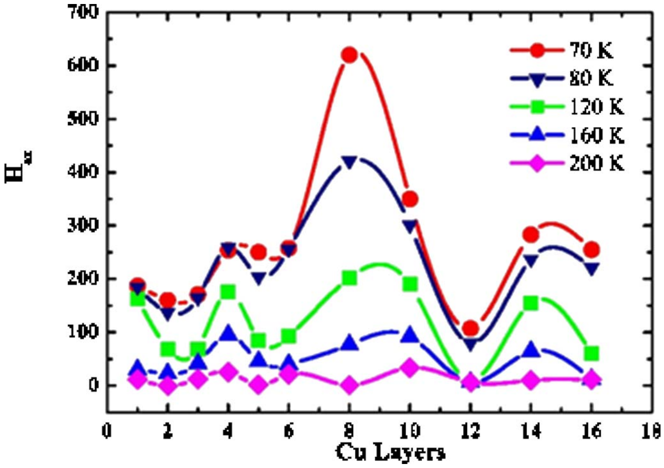

FIG. 3. (Color online) Exchange bias vs. Cu spacer thickness for samples with variation of temperature to be 70, 80, 120, 160, and 200 K.

amplitude decreases as the temperature increases up to the blocking temperature. This monotonic variation of the oscillation amplitude with respect to the temperature is vividly demonstrated in Fig. 3. On the other hand, it is very interesting to note that the oscillation amplitude *with respect to the Cu spacer layer thickness* becomes nonmonotonic as depicted in the figure. The oscillation amplitude initially increases as the Cu spacer layer thickness (N_{Cu}) increases. It reaches the maximum when $N_{Cu}=8-10$ for all the measurement temperatures and decays again for the sample with thicker Cu spacer layers. The nonmonotonic variation of oscillation amplitude in interlayer exchange coupling sounds quite striking compared to the relatively simple case of FM/NM/FM trilayer, where monotonic decreasing of oscillation amplitude with respect to the thickness is expected. This interesting feature of nonmonotonic variation of oscillation amplitude in the interlayer exchange coupling opens a possibility of oscillation amplitude controllability with variation of nonmagnetic spacer layer thickness in AFM/NM/FM trilayer system. Initial increase of the oscillation amplitude with increase of Cu spacer thickness, leading to the overall nonmonotonic amplitude variation, has been theoretically studied in the Ref. 18, where the oscillation amplitude variation has been investigated with the consideration of coupling contribution from extended states across the nonmagnetic spacer layer. However, the exact mechanism that generates the nonmonotonic variation of oscillation amplitude requires a further investigation.

In conclusion, we report our experimental finding that there exists an oscillatory interlayer exchange coupling in CoO/Cu/Co trilayer with respect to the Cu spacer layer thickness variation. The oscillation period about four Cu layer thickness seems to be irrespective of the temperatures within our measurement error. Very interestingly, the oscillation amplitude exhibits nonmonotonic behavior with respect to the Cu spacer layer thickness. It initially increases, reaches the maximum, and then decreases as the Cu spacer thickness increases, which implies a possibility of controlling the oscillatory interlayer exchange coupling behavior.

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