# Oscillatory Interlayer Exchange Coupling and Its Temperature Dependence in $[Pt/Co]_3/NiO/[Co/Pt]_3$ Multilayers with Perpendicular Anisotropy

Z. Y. Liu and S. Adenwalla

*Department of Physics and Astronomy, University of Nebraska-Lincoln, Lincoln, Nebraska 68588-0111, USA*

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Interlayer exchange coupling that oscillates between antiferromagnetic and ferromagnetic as a function of NiO thickness has been observed in [Pt(5 Å)/Co(4 Å)]$_3$/NiO($t_{\mathrm{NiO}}$ Å)/[Co(4 Å)/Pt(5 Å)]$_3$ multilayers with out-of-plane anisotropy. The period of oscillation corresponds to $\sim$2 monolayers of NiO. This oscillatory behavior is possibly attributed to the antiferromagnetic ordering in NiO. The antiferromagnetic interlayer exchange coupling for the 11 Å NiO layer shows an increase in coupling strength with increasing temperature, in agreement with the quantum interference model of Bruno for insulating spacer layers. A coexistence of exchange biasing and antiferromagnetic interlayer exchange coupling has been observed below $T = 250$ K.

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Ferromagnetic (FM) layers separated by nonmagnetic metallic spacers display oscillatory interlayer exchange coupling (IEC) between the FM layers. This behavior appears to be a rather general feature and has been seen in a large variety of materials [1–3]. The period of oscillation is related to spanning vectors of the Fermi surface of the spacer material lying perpendicular to the layer plane [4,5], and the coupling arises from a RKKY type coupling. In carefully grown Fe/Cr/Fe sandwiches [6], two oscillatory periods are visible: a short one corresponding to 2 monolayers (MLs) of Cr and a long one corresponding to 18 MLs of Cr. The antiferromagnetic (AF) ordering of Cr is stabilized by the presence of the FM Fe [7]. For insulating spacer layers, nonoscillatory decay of the IEC strength with spacer thickness has been observed [8,9], and the IEC strength increases with temperature [10] in contrast to the case for metallic spacers. Generalization of the IEC theory to insulating spacers by introducing a complex Fermi surface [11–13] suggests that the IEC strength (either FM or AF) should show a nonoscillatory decay with the spacer thickness and increase with temperature, being consistent with the experimental observation. Recently, a coexistence of exchange biasing and FM coupling in two FM layers with inplane anisotropy separated by a NiO layer has been reported [14]. In the majority of these coupled multilayers, the magnetization lies in the plane of the film. However, there have been reports of out-of-plane exchange coupling in Co/Ru [15,16] and Ni/Cu [17] superlattices. Observations of the IEC in FM/insulating spacer/FM systems with perpendicular anisotropy are scarce.

In this Letter we have investigated the IEC at room temperature (RT) as a function of the NiO thickness in glass /Pt(100 Å)/[Pt(5 Å)/Co(4 Å)]$_3$/NiO($t_{\mathrm{NiO}}$ Å)/[Co(4 Å)/Pt(5 Å)]$_3$/Pt(50 Å) multilayers with an out-of-plane easy axis. The temperature dependence of the coupling strength for 11 Å NiO has also been studied.

The samples were prepared by dc and rf magnetron sputtering from separate Pt, Co, and NiO targets at the deposition rates of 0.96, 0.2, and 0.19 Å/s, respectively, in 3 mTorr Ar pressure. The base pressure was $4 \times 10^{-7}$ Torr. The entire set of samples with different thicknesses of NiO were grown in the same run and at the same conditions. Two additional samples of glass/Pt(100 Å)/[Pt(5 Å)/Co(4 Å)]$_3$/NiO(11 Å) (S1) and glass/NiO(11 Å)/[Co(4 Å)/Pt(5 Å)]$_3$/Pt(50 Å) (S2) were also grown in the same run. The thickness calibration was checked by grazing angle x-ray reflectivity after sample preparation, displaying an accuracy of $\sim$10%. X-ray diffraction results show highly textured fcc(111) Pt and NiO layers and hcp(100) Co layers.

Hysteresis $M - H$ loops have been measured by a superconducting quantum interference device with field applied perpendicular to the sample surface. The $M - H$ loops at RT for the samples of S1 and S2 show that the coercivity (887 Oe) of the former is much higher than the latter (80 Oe), indicating that, for the two Co/Pt multilayers separated by a NiO layer, the lower multilayer is magnetically hard while the upper one is magnetically soft. Thus, the IEC can be obtained from the minor loop shift of the upper multilayer, while the lower multilayer remains pinned [9,18,19]. The hysteresis loops of these as-prepared samples do not display any shift at RT.

Figure 1(a) depicts the major and minor $M - H$ loop at RT for NiO thickness of $t_{\mathrm{NiO}} = 11$ Å. The minor loop displays a large net positive shift of 619 Oe. This shift does not result from the exchange biasing (EB) due to the AF NiO layer, because the EB disappears completely above 250 K (as discussed later in the Letter). It can be unambiguously attributed only to the IEC between the two multilayers across the thin NiO spacer. Magnetic force microscopy (MFM) imaging in Fig. 1(b) demonstrates the existence of 360° domain walls, identical to those observed in the AF coupled Co/Ru/Co multilayers

  

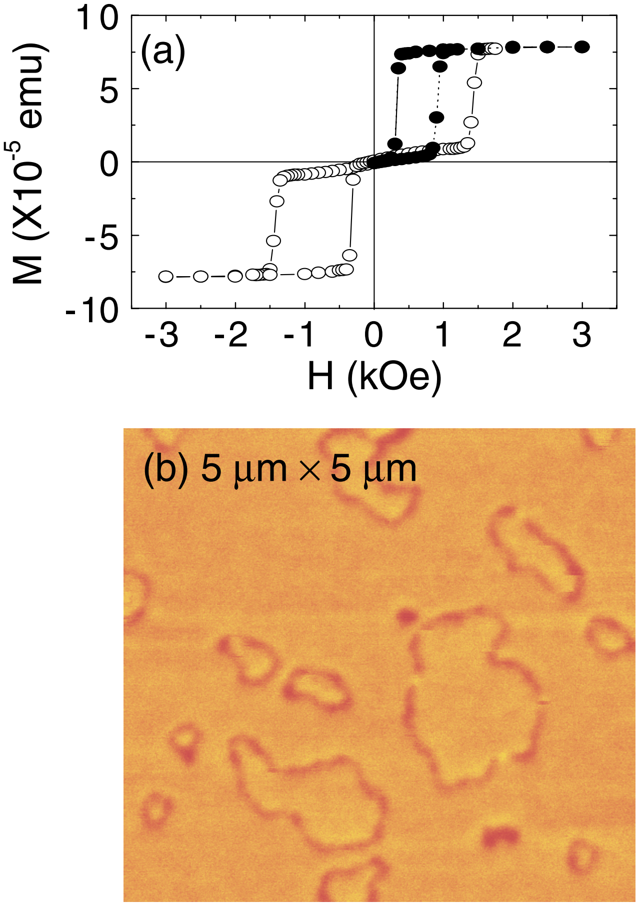


FIG. 1 (color online). (a) The major (○) and minor (●) $M-H$ loops along the out-of-plane easy axis for glass/Pt(100 Å)/[Pt(5 Å)/Co(4 Å)]$_3$/NiO(11 Å)/[Co(4 Å)/Pt(5 Å)]$_3$/Pt(50 Å). The minor loop is measured after a positive saturation of the whole system, in a field range where the magnetically hard lower multilayer is magnetically fixed. In this sample, the large shift of 619 Oe indicates a strong AF coupling across the 11 Å NiO layer. (b) The magnetic force microscopy image at the remanent state ($H = 0$ Oe).

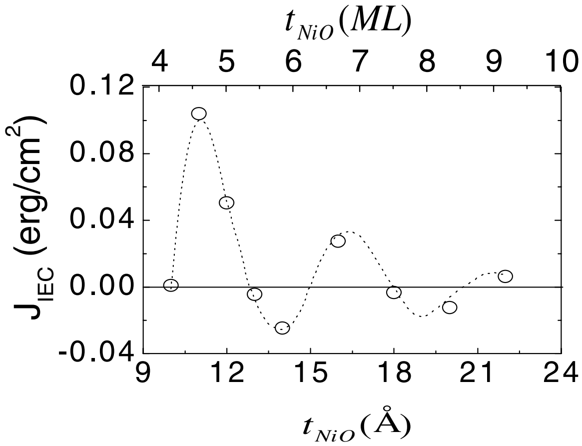


FIG. 2. The IEC strength ($J_{IEC}$) as a function of NiO thickness (in units of both Å and ML) at room temperature. $J_{IEC}$ is determined by where $M_S$ and $t_F$ are the saturation magnetization and total thickness of all upper Co layers, respectively; $H_{MLS}$ is the minor loop shift with $H_{MLS} > 0$ ($< 0$) corresponding to the AF (FM) coupling. The dotted line is a guide to the eyes.

with perpendicular anisotropy [16], corroborating that the minor loop shift at RT is due to the AF coupling.

The IEC strength $J_{IEC}$ across the NiO layer at RT is determined by the minor loop shift $H_{MLS}$ through the expression $J_{IEC} = H_{MLS} M_S t_F$, where $M_S$ and $t_F$ are the saturation magnetization and the total thickness of all Co layers in the upper Co/Pt multilayer. Figure 2 shows clearly that at RT the IEC oscillates between AF and FM coupling as a function of NiO thickness with a period of ~5 Å. This unexpected oscillatory behavior is quite different from the nonoscillatory decay of the IEC strength expected by the models of Bruno [4,13] and Slonczewski [11] for nonmagnetic insulating spacers and from recent experimental observations of coupling across a nonmagnetic insulating MgO spacer [9]. The insulating spacer is modeled by a rectangular potential barrier height of $U_0$ higher than the Fermi level $E_F$ of the FM layers, and the IEC is ascribed to the interference of electron waves in the spacer layer due to spin-dependent reflections at the FM/insulator interfaces [4,13]. However, the NiO layer is not only an insulator but also an AF material. Experimental studies [20] have shown a Néel temperature of 295 K for NiO films as thin as 5 MLs. The AF ordering of an AF layer in contact with a FM layer will be stabilized [21]. Hence, for the NiO layers in our samples, they should have a Néel temperature higher than RT. Taking the AF ordering of NiO into consideration, we propose a possible explanation for the oscillatory IEC seen in our system.

The NiO layer in our samples is highly (111) textured. In bulk NiO, the spins lie in ferromagnetically ordered (111) planes with (111) planes stacking antiferromagnetically. Assuming a similar spin structure in our thin NiO film, successive (111) planes of NiO will have net magnetizations pointing in opposite directions, lying in the plane of the sample. The magnetization of each (111) plane will contribute to the scattering of spin-polarized electron waves from the FM (111) planes. The presence of AF domains, interfacial roughness, and different inplane crystalline orientations in the NiO layer will alter the magnitude of the net magnetization, but the AF ordering of the NiO (111) planes ensures that electron waves traveling through the barrier will experience magnetic fields that are opposite in direction for each successive NiO (111) plane. Rather than a rectangular potential barrier with a width given by the spacer thickness [11], one may consider a periodic potential $V(x)$ with a period of $2d$ inside the NiO layer [where $d = 2.41$ Å is the distance between (111) planes]. For simplicity, $V(x)$ can be considered to be composed of periodically arranged rectangular carriers similar to the atomic plane model for the metallic spacer by Bruno *et al.* [22]. This periodic rectangular potential barrier will cause multiple reflections of electron waves from the FM ordered (111) planes in the NiO layer as well as the Co/NiO interfaces. Their interference may then allow for a modulation of the reflectivity through the NiO by a function which is periodic in the NiO thickness with a period of $2d = 4.8$ Å, which is consistent with the oscillatory period of $J_{IEC}$. This oscillatory behavior of the IEC as a function of NiO thickness is different from the oscillation in Fe/Cr multilayer as a function of Cr thickness. Even though Cr is an antiferromagnet, it is metallic. NiO is an antiferromagnetic insulator, and the oscillatory IEC is very likely to originate from the antiferromagnetism of NiO spacers.

In the above explanation, the out-of-phase magnetization does not play a significant role, since, according to the model by Slonczewski [11], it is only the relative orientation of magnetization in the FM layers that is important, which in our case is either parallel or antiparallel to the out-of-plane easy axis.

We have studied the temperature dependence of the strongest AF coupling strength across the 11 Å NiO layer. This sample is cooled at zero field from 300 to 30 K. After cooling, a series of major and minor $M - H$ loops have been measured at different temperatures while warming up to RT. Figure 3 shows the loops at a few representative temperatures. Below 250 K, the upper Co/Pt multilayer is shifted towards the positive field direction, but the lower one is shifted towards the negative field direction. These shifts of the major $M - H$ loops are due to the EB effect of the AF NiO layer, because the IEC across the NiO layer alone cannot cause any net shift of the major $M - H$ loop. The exchange fields for the upper ($H_{UE}$) and lower ($H_{LE}$) multilayers have been extracted from the upper and lower shifts of the $M - H$ loops, respectively, and their temperature dependences are shown in Fig. 4(a). The opposite signs are consistent with the fact that at remanence ($H = 0$) the magnetizations of the two Co/Pt multilayers are oppositely directed. Both $H_{UE}$ and $H_{LE}$ decrease linearly with temperature and disappear at a blocking temperature of $T_B = 250$ K. This linear temperature dependence of the exchange fields can be explained by Malozemoff's model for exchange-biased FM/AF bilayers [23]. Experimental studies have shown that the appearance of exchange bias is strongly related to domains in the AF layer [24–26]. Although, theoretically, domain walls parallel or perpendicular to the interface are possible [23,27], for our very thin NiO layer only perpendicular domain walls are feasible. With the assumption that $H_E \propto \sqrt{A_{AF}K_{AF}}$, the domain wall energy (where $A_{AF}$ and $K_{AF}$ are the exchange constant and the magnetocrystalline anisotropy of the AF material, respectively), it has been shown that $H_E \propto \sqrt{A_{AF}K_{AF}(0)}(1 - T/T_B)$ for AF materials with cubic anisotropy [23,28], which is consistent with our data.

The minor loops as shown in Fig. 3 are always shifted towards the positive field direction, and the minor loop shift $H_{MLS}$ represents the total coupling strength $J_{MLS}$

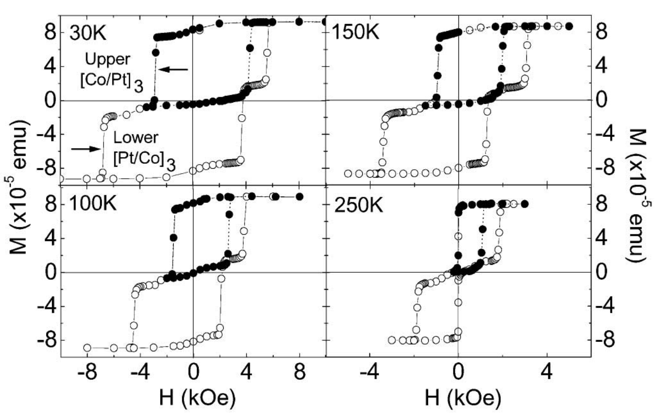


FIG. 3. The major (○) and minor (●) $M - H$ loops along the out-of-plane easy axis for glass/Pt(100 Å)/[Pt(5 Å)/Co(4 Å)]$_3$/NiO(11 Å)/[Co(4 Å)/Pt(5 Å)]$_3$/Pt(50 Å) at some typical temperatures.

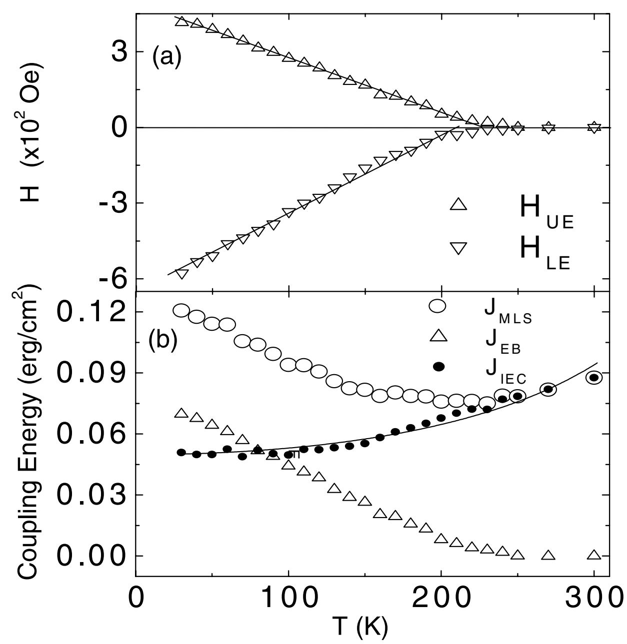


FIG. 4. (a) Temperature dependences of the exchange fields for the upper ($H_{UE}$) and lower ($H_{LE}$) multilayers of glass/Pt(100 Å)/Pt(5 Å)/Co(4 Å)]$_3$/NiO(11 Å)/Co(4 Å)/Pt(5 Å)]$_3$/Pt(50 Å). The solid lines are the linear fit to the data according to Malozemoff's model [23]. (b) The temperature dependences of the total coupling energy $J_{MLS}$ (○), the exchange biasing energy $J_{EB}$ (△) for the upper Co/Pt multilayer, and the IEC energy $J_{IEC}$ (●). The solid line is the fit to the $J_{IEC} - T$ curve according to $J_{IEC}(T) = J_{IEC}(0)x/\sin x$ (for definitions of the parameters, see the text).

across the NiO layer by $J_{\rm MLS} = H_{\rm MLS} M_S t_{\rm F}$. The temperature dependence of $J_{\rm MLS}$ is shown in Fig. 4(b) together with the EB energy $J_{\rm EB}$ determined by $J_{\rm EB} = H_{\rm UE} M_S t_F$. $J_{\rm MLS}$ is much higher than $J_{\rm EB}$, suggesting that $J_{\rm MLS}$ is determined not only by $J_{\rm EB}$, but also by the contribution of the IEC energy $J_{\rm IEC}$ across the NiO layer. Thus, $J_{\rm MLS}$ can be expressed as $J_{\rm MLS} = J_{\rm EB} + J_{\rm IEC}$. The temperature dependence of $J_{\rm IEC}$ is shown in Fig. 4(b). It can be seen clearly that $J_{\rm IEC}$ increases with increasing temperature, being consistent with the quantum interference model of Bruno for insulating spacers [4,13], in which the temperature dependence of the IEC is controlled by $J_{\rm IEC}(T) = J_{\rm IEC}(0)x/\sin x$, where $J_{\rm IEC}(0)$ is the IEC strength at $T = 0$ K and $x = 2\pi k_B T t_{\rm NiO} m/\hbar^2 k_F$. $m$ is the electron mass, $k_B$ is Boltzmann's constant, $\hbar$ is Planck's constant, and $k_F$ is the complex Fermi wave vector of the NiO spacer. A fit to the data gives the values of $J_{\rm IEC}(0) = 0.049$ erg/cm$^2$ and $k_F = 0.14$/Å. The good agreement shown in Fig. 4 suggests that Bruno's model provide a plausible explanation for the experimental observation of the IEC coupling across the NiO layer. Experimental studies [29,30] have demonstrated that NiO is a charge-transfer insulator and the electron mobility in NiO can be highly thermally activated with increasing temperature. Hence, the increase of the IEC strength with temperature is induced by thermally activated electron mobility in the NiO layer as suggested by Bruno's model for an insulating spacer. The temperature dependences of the IEC strength and the exchange bias provide strong confirmation that even at a thickness of just 11 Å, the NiO layer still preserves the characteristics of an antiferromagnetic insulator.

In summary, oscillatory interlayer exchange coupling has been observed at RT in $[\mathrm{Pt}(5\ \text{Å})/\mathrm{Co}(4\ \text{Å})]_3/\mathrm{NiO}(t_{\rm NiO}\ \text{Å})/[\mathrm{Co}(4\ \text{Å})/\mathrm{Pt}(5\ \text{Å})]_3$ multilayers with out-of-plane anisotropy. The unexpected oscillatory behavior of the IEC as a function of NiO thickness is thought to be related to the AF ordering of the NiO layer. The increase of the AF coupling strength with temperature can be well understood by the quantum interference model of Bruno [4,13] and confirms that the IEC across the insulating NiO layer is thermally induced. A coexistence of exchange biasing and AF interlayer exchange coupling has been observed after cooling the sample to 30 K at zero field. The linear temperature dependences of the exchange bias follow from the cubic anisotropy of the NiO layer due to the formation of perpendicular domain walls in the NiO layer as proposed by Malozemoff [23].

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