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Temperature-dependent crossover from ferro- to antiferromagnetic interlayer alignment due to magnetic anisotropy energy

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The temperature dependence of the interlayer coupling of perpendicularly magnetized Ni/Cu/Ni(001) trilayers is studied. A crossover in the alignment of the sublayer magnetizations from parallel to antiparallel is observed with increasing temperature, although the interlayer exchange is measured to be antiferromagnetic for all temperatures. The crossover is the result of the competition between the antiferromagnetic interaction and the magnetic anisotropy energy. This Ni/Cu/Ni trilayer with moderate exchange coupling serves as a model for magnetic multilayers in which the interlayer coupling can be switched with temperature.
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The oscillatory interlayer exchange coupling of ferromagnetic transition metal layers through nonmagnetic spacers has been intensely studied over the last decade. Its dependence on the thickness of the spacer and the ferromagnetic layers and on the crystallographic orientation in tri- and multilayers has been experimentally determined and theoretically described.[1–7] In comparison, relatively little work was devoted to the temperature dependence of the exchange coupling. Exchange coupling *a priori* is temperature independent. In multilayers, however, the interlayer coupling follows a monotonic temperature dependence,[8–11] which is attributed to the softening of the density of states at the Fermi edge.[7]

In this model a change of sign of the coupling constant J is not expected. Experimentally, however, a crossover from ferromagnetic (FM) to antiferromagnetic (AFM) orientation of the layer magnetizations was observed with decreasing[8] or increasing[9,10] temperature. This was attributed to sample imperfections leading to a competition between AFM and FM coupled regions. Here we would like to draw attention to a fundamental mechanism which has not been discussed appropriately in the literature of exchange-coupled multilayers, that is, the importance of orbital momentum and magnetic anisotropy energy (MAE) for the ordering of the layer magnetizations in weakly coupled multilayers. In limiting cases MAE has been included in the discussion of the reversal process of the magnetization in coupled layers resulting in simple models where MAE or J could be used as fitting parameters only.[12] The temperature dependence of *both* quantities was not considered. Here we focus explicitly on the temperature dependence of J and MAE. It is well known that most thin film structures are distorted, for example, tetragonal Ni/Cu(001),[13–17] which lifts the quenching of the orbital momentum and gives rise to a by-orders-of-magnitude-increased MAE. The latter one becomes of similar magnitude as the exchange constant, and it is temperature dependent. We suggest that MAE should be taken into account and that not only J but also MAE determines the effective interlayer coupling, and it serves to manipulate the coupling phenomena for multilayer application as we will show in this paper.

We demonstrate experimentally that in a perpendicularly magnetized 12 ML Ni(001)/5 ML Cu(001)/9 ML Ni(001) trilayer deposited on a Cu(001) single crystal the remanent sublayer magnetizations change from FM to AFM ordering with increasing temperature. Despite this change in ordering we find that the interlayer exchange coupling is antiferromagnetic at all temperatures and decreases linearly with temperature in the expected way.[7,11] The origin of this apparent crossover from FM to AFM originates from orbital magnetism and the resulting MAE. At low temperature MAE dominates and fixes the sublayer magnetizations at remanence in a parallel orientation after application of an external field $H = 100$ Oe. With increasing temperature the MAE decreases more rapidly than the AFM interlayer exchange and the remanent state of the trilayer flips to an AFM alignment. From these results based on a careful analysis of minor and major hysteresis loops we conclude: (a) The determination of the exchange-coupling constant sign from the remanent magnetization state measured, for example, in electron spectroscopies[1,8,10] may give the wrong sign of J in weakly coupled multilayers. (b) The consequences of orbital momentum, that is spin-orbit coupling and MAE as a function of temperature need to be included in the theory of coupling phenomena. (c) Manipulation of the MAE may provide a useful tool to tune the FM-to-AFM crossover temperature for technological application.

The thickness and temperature dependencies of the MAE of Ni/Cu(001) ultrathin films have been thoroughly studied.[13–15,18] Up to seven monolayers (ML), the easy axis of the magnetization lies in the film plane and an unusual spin-reorientation phase transition to the out-of-plane direction occurs by increasing the film thickness.[18] The temperature dependence of the magnetization and its specific features near the Curie temperature (T_C) for perpendicularly magnetized Ni/Cu(001) films are also available.[14] In the present work, a 9-ML-thick Ni film was prepared on a Cu(001) substrate under ultrahigh vacuum (UHV) as described elsewhere.[13–16] Ni on Cu(001) grows layer by layer in a tetragonal face-centered symmetry with very small interface roughness after annealing to 450 K.[16,17] On the top of the first Ni layer, a 5 ML Cu layer with a thickness gradient of 0.1 ML/mm was grown. This Cu thickness is close to the crossover between ferro- and antiferromagnetic coupling for

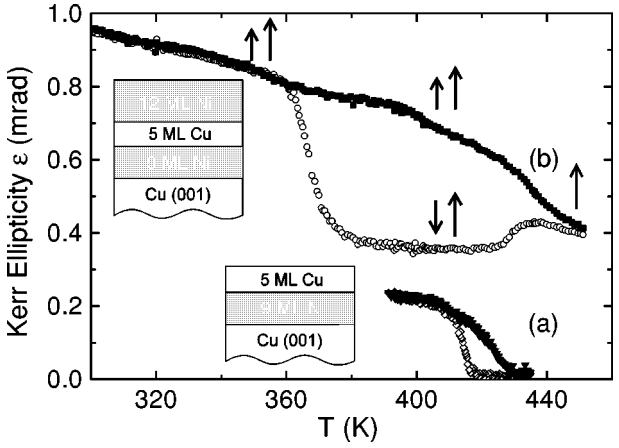

FIG. 1. Kerr ellipticity at remanence (○, ◇), at 50 Oe (▼) and at 100 Oe (■) for 5 ML Cu/9 ML Ni/Cu(001) (a) and for the trilayer (b) as a function of temperature. The orientations of the magnetization of the bottom (short) and the top layer (long arrow) are indicated.

various transition metals (Fe, Co, Ni) separated by Cu(001).[19] Finally, a 12 ML Ni layer was grown on the top of the bilayer.

The magnetic properties of the trilayers at temperatures higher than room temperature (RT) were studied *in situ* via the polar magneto-optic Kerr effect (MOKE). Measurements were performed along the easy axis of magnetization. Temperature-dependent magnetization curves were recorded in a way described earlier.[14] Over many heating cycles up to 450 K no changes in the magnetic response were observed.

In Fig. 1(a) the perpendicular Kerr ellipticity at remanence ε_r and at an external field $\varepsilon(H=50\ \mathrm{Oe})$ is shown for the 5 ML Cu/9 ML Ni/Cu(001) bilayer. Similar behavior near the Curie temperature was observed in our previous studies on perpendicularly magnetized 8–10 ML Ni/Cu(001) ultrathin films.[14] The ε_r and $\varepsilon(H=100\ \mathrm{Oe})$ data for the trilayer 12 ML Ni/5 ML Cu/9 ML Ni/Cu(001) between RT and 450 K are shown in Fig. 1(b). The total Kerr ellipticity signal ε is strongly increased due to the contribution of the top Ni layer. Up to 360 K ε_r is equal to $\varepsilon(H=100\ \mathrm{Oe})$ and the sublayer magnetizations are parallel. Above 360 K ε_r is abruptly reduced and the sublayer magnetizations are antiparallel at $H=0$ Oe and parallel at $H=100$ Oe as the arrows indicate. ε_r coincides again with $\varepsilon(H=100\ \mathrm{Oe})$ at higher temperatures (close to 450 K), where the magnetization of the bottom Ni layer has vanished after entering the paramagnetic phase. This behavior is observed reversibly over the full temperature interval when heating and cooling several times. The signal at 450 K originates exclusively from the top Ni layer. In a simple analysis one could conclude that the exchange coupling has reversed from FM to AFM near a crossover temperature $T_x \approx 370$ K. However, the results presented below unambiguously show that also in the FM coupled region ($T<370$ K) the exchange is AFM!

For the trilayer shown in Fig. 1 hysteresis loops were recorded at various temperatures. In Fig. 2(a) the RT hysteresis loop is plotted. The field of 100 Oe forces both sublayer magnetizations (M_{bot} for the bottom and M_{top} for the top Ni layer) to be parallel to each other. At $H\approx -30$ Oe the bottom layer magnetization (smaller arrow) starts to reverse and at

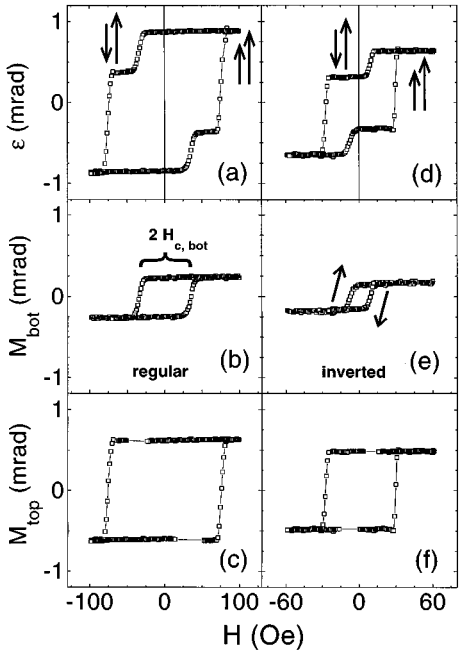

FIG. 2. Hysteresis loops at RT (a)-(c) and 396 K (d)-(f) as explained in the text. Arrows in (a) and (d) indicate the orientations of the two sublayer magnetizations. The arrows in (e) show the path of the bottom layer magnetization.

$H=-50$ Oe an antiparallel configuration of the bottom and top layer magnetization is established. At about -75 Oe also M_{top} starts to reverse towards the external field, and the process is completed just below the field of -100 Oe. From the hysteresis loop of Fig. 2(a) it is easy to separate the contributions of M_{bot} [Fig. 2(b)] and M_{top} [Fig. 2(c)]. The experimental hysteresis loops and the extracted bottom and top layer loops at $T=396$ K are shown in Figs. 2(d)-2(f). The main difference between 303 K and 396 K is the orientation of the sublayer magnetizations M_{bot} at remanence [Figs. 2(a) and 2(d)]. The extracted minor loop [Fig. 2(e)] shows that M_{bot} follows an inverted path above 360 K. Near remanence M_{bot} at 396 K is oriented opposite to the external field at 396 K while at 303 K the regular behavior is found. To show more clearly the existence of AFM interlayer exchange coupling at *both* temperatures we traced minor-hysteresis loops in the following way: The orientation of M_{top} was kept fixed since its coercivity was not exceeded when recording the minor loops shown in Fig. 3. The asymmetry of the top and bottom layer made it possible to trace the loop of M_{bot} only, see, for example, Ref. 20. The results at RT (open squares) and 396 K (closed squares) are shown in Fig. 3. In both cases the minor loops are exchange shifted towards the same direction and the negative sign of H_{exch} indicates an antiferromagnetic interaction. The H_{exch} values of 17 Oe at 396 K and 32 Oe at RT correspond to values of the interlayer exchange coupling constant $J<10^{-3}$ erg/cm^2 (0.4 μeV/atom).[6] These values are more than two orders of magnitude smaller than for most tri- or multilayers.

The temperature dependence of the exchange field H_{exch} is plotted in Fig. 4. A linear dependence of H_{exch} (closed squares) as a function of temperature is observed which is in

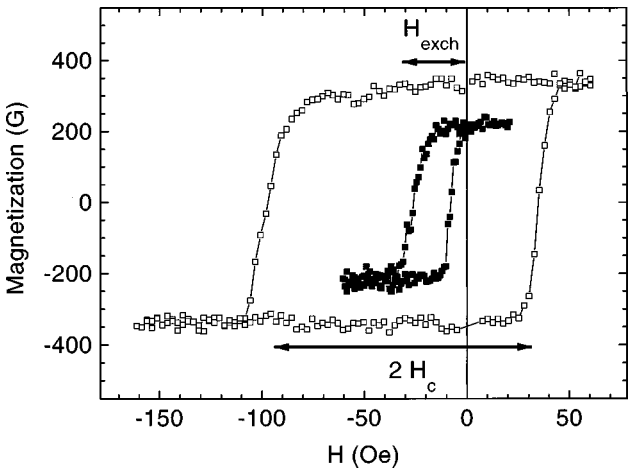

FIG. 3. Minor-hysteresis loops showing the magnetization of the bottom layer at 303 K (open squares) and 396 K (solid squares). M_{top} was biased along the negative field direction. The negative exchange field H_{exch} indicates the antiferromagnetic interaction. The magnetization has been calibrated in Gauss as explained in Ref. 14.

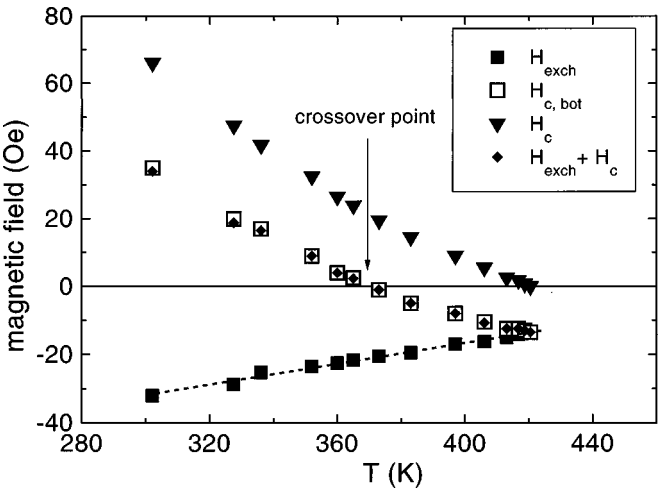

FIG. 4. H_{exch} (closed squares), H_c (closed triangles), $H_{c,\text{bot}}$ (open squares) or $(H_{\text{exch}}+H_c)$ (closed diamonds) as a function of temperature. The dashed line is a guide to the eye.

agreement with reports on other exchange-coupled multilayers.[7,11] No change of the sign of H_{exch} and thus of the interlayer exchange with temperature is observed. The values of the coercivities of the minor loops H_c (Fig. 3) are also included in Fig. 4 (closed triangles), together with the values of $H_{c,\text{bot}}$ [Figs. 2(b) and 2(e)] (open squares). One should note that the temperature dependence of H_c of the minor loop is related to the one of MAE (Ref. 21) which has been measured previously.[13,15,18] It decreases more strongly than the exchange field, and the competition between H_c, that is MAE, and H_{exch}, that is exchange coupling, determines the orientation of the magnetizations. This is even more evident if one regards the sum of $H_{\text{exch}}+H_c$ (diamonds) which is identical with the coercivity $H_{c,\text{bot}}$ of the bottom layer (Fig. 4). Hence, we conclude that the change of the sign of $H_{c,\text{bot}}$, that is the crossover to AFM coupling, at 360–370 K is the result of this competition. Thus the effects of the antiferromagnetic exchange coupling favoring an antiparallel sublayer magnetization alignment become more prominent at higher temperatures, and a coupling crossover is recorded. However, the interlayer exchange is AFM at all temperatures. This is an important result since it shows that the balance of MAE *and* AFM exchange as a function of temperature determines the reorientation of the bottom layer.

The problem to determine the exchange coupling sign from the remanent magnetization was also discussed for example by Heinrich *et al.*[22] but only at a single temperature. They observed a parallel sublayer remanent magnetization while the interlayer coupling was found to be antiferromagnetic. Also recent magnetoresistance data showed the effect of MAE.[23] However, the temperature dependence of MAE was not experimentally determined, and the anisotropy constants were used as fit parameters only.

In this work we have measured a crossover from ferromagnetic to antiferromagnetic interlayer order as a function of temperature in weakly coupled trilayers. We show that it is an apparent crossover of the coupling due to the temperature-dependent competition of the antiferromagnetic interlayer exchange coupling and the magnetic anisotropy energy. The exchange coupling is antiferromagnetic at *all* temperatures. The large MAE dominates at low temperature and fixes the sublayer magnetizations in a parallel configuration at remanence. At higher temperature the MAE is strongly reduced and the AFM exchange coupling dominates yielding the experimentally observed AFM configuration at remanence. It is concluded that orbital momentum and MAE which are strongly enhanced in noncubic multilayer structures should be included in the analysis of coupling crossover phenomena. Last, but not least, we mention that our crossover temperature T_x of 370 K is in a perfect range for technological application. Small manipulation in metallurgy and film preparation may bring T_x to slightly above ambient temperature which is best for practical use.

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