Vanishing Magnetic Interactions in Ferromagnetic Thin Films

J. Hunter Dunn,[1] O. Karis,[2] C. Andersson,[2] D. Arvanitis,[2] R. Carr,[3] I. A. Abrikosov,[4]
B. Sanyal,[2] L. Bergqvist,[2] and O. Eriksson[2]
[1]*MAX-lab, Box 118, S-221 00, Lund, Sweden*
[2]*Department of Physics, Uppsala University, Box 530, 751 21 Uppsala, Sweden*
[3]*Stanford Synchrotron Radiation Laboratory, SLAC, 2575 Sand Hill Road, Menlo Park, California 94025, USA*
[4]*Department of Physics and Measurement Technology, Linköping University, SE-58183 Linköping, Sweden*
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We have used element-specific hysteresis measurements, based on the x-ray magnetic circular dichroism technique, to investigate magnetic trilayer structures composed of Fe and Ni layers. Within a critical regime we have discovered a class of structures in which the exchange interaction, the mechanism responsible for the macroscopic magnetism, can become vanishingly small. The experimental observations are supported by first principles theory and are explained as arising from a cancellation of several competing magnetic interactions. Hence, we have discovered a system with a novel exchange interaction between magnetic layers in direct contact that replaces the conventional exchange interaction in ferromagnets.

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The coupling between atomic spins in solids is provided by exchange interactions. Several mechanisms have been proposed to explain such phenomena. Examples include the double exchange, the super exchange, the RKKY interaction, direct exchange, and the interlayer exchange. The latter two forms of exchange interaction are of particular interest because of their importance in information technology [1]. Indeed, the discovery of interlayer exchange interaction [2–4] found in metallic multilayers, that is, the interaction which mediates the magnetic coupling of two magnetic films over a nonmagnetic spacer layer, has led to intense research efforts in the area of thin film magnet structures [5]. Typically, the systems considered are layered structures that are composed of magnetic materials separated by layers of a nonmagnetic material, or *spacer layers* (e.g., V or Cu). Typical values for the strength of the interlayer exchange energy are of order 1–1000 $\mu J/m^2$, which is 3–5 orders of magnitude smaller than the direct exchange interaction. Here we report the discovery of a novel coupling behavior between magnetic layers (Fe and Ni) in direct contact arising from a cancellation of interaction terms leaving an interaction which is several orders of magnitude weaker, i.e., of the same strength as the interlayer exchange found across a nonmagnetic spacer layer. Hence, we have discovered a system that, despite the absence of a spacer layer, exhibits a magnetic decoupling of the constituent Fe and Ni layers.

The growth of Fe or Ni ultrathin films on the Cu(001) substrate has been investigated thoroughly in the past. The films are reported to grow in a metastable layer-by-layer mode, with a face-centered-cubic (fcc) structure with varying degree of tetragonal distortion [6,7]. In the ultrathin limit, the two systems exhibit different orientations of the easy magnetization direction when grown on Cu(001). Below 4 atomic layers (AL) Fe exhibits an out-of-plane anisotropy, while Ni exhibits an in-plane anisotropy up to about 8 AL. Although layer-by-layer fcc growth has been demonstrated, it is also clear that some intermixing of the Fe and Ni atoms is probable. Investigations of Fe/Ni bilayer structures grown on Cu(001) have been reported previously [8,9].

The experiments were performed at beam line 5.2 of the Stanford Synchrotron Radiation Laboratory (SSRL) and at beam line D1011 of MAX-lab. These beam lines provide soft x rays covering the L absorption edges of the magnetic $3d$ transition metals with a variable state of polarization. Spectroscopies performed at these edges can, with the proper choice of x-ray polarization and experimental geometry, provide information on the magnetic state of the sample with elemental specificity [10].

A total of nine sample configurations of the form $Ni_pFe_qNi_r/Cu(001)$ were grown by subsequent evaporations of each element; $p = 8$–15 AL, $q = 2$–3 AL, and $r = 2$–3 AL. The magnetic properties were characterized using an element-specific x-ray magnetic circular dichroism (XMCD) technique and soft x-ray resonant reflectivity or x-ray resonant magnetic scattering. To ensure the highest possible quality, all samples were prepared and measured *in situ* in ultrahigh vacuum with a base pressure better than 2×10^{-8} Pa. The films were prepared by evaporation of purified bulk Fe and Ni, a standard procedure used to produce high quality films. The film thickness was calibrated *in situ* using the method described in Ref. [11]. Throughout the experiments, the sample was maintained below 150 K apart from a short annealing to 300 K after film deposition. This method has been reported to result in an increased ordering of the individual layers while having a negligible impact on the magnetic properties of the constituent layers [7,12]. The measurements were performed at 20–30 K. For each stage of preparation

the magnetic properties of the constituent layers were characterized.

In this Letter we focus on the particular structure $Ni_2Fe_2Ni_8/Cu(001)$ which exhibits the most striking decoupling phenomena [13]. We found that the initial evaporation of 8 AL of Ni results in an in-plane easy direction of the magnetization. The addition of 2 AL of Fe turns the easy direction out of plane. Finally, the last two layers of Ni turns the easy direction back in plane. This behavior is in agreement with the combined anisotropy energy of the bilayer and trilayer, as calculated using the surface and volume contributions of the individual Fe and Ni films grown separately on Cu(100) [14,15]. This confirms that the structure of the Fe and Ni layers of our trilayer was not markedly different from individual ultrathin films grown on Cu(100). We conclude that our trilayer system did not correspond to a random FeNi alloy, leaving distinct, albeit roughened, interfaces as the likely situation. This conclusion is further supported by characteristic intensity variations in the x-ray absorption data.

We used the element-specific aspect of the technique to obtain independent magnetic hysteresis loops from the constituent Ni and Fe atoms in the trilayer system. The most striking result, shown in Fig. 1, is that the coercive field of the Fe and Ni layers is different and that the Fe and Ni layers reverse, leading to a nonparallel coupling for certain points in the cycle. The overall squareness of the hysteresis loops suggests that each constituent layer is close to a monodomain. Starting from a negative applied field, with the system in saturation (in this case -1.2 kA/m or -15 Oe), as we reduce the field toward zero the coupling between Fe and Ni remains parallel. However, as we pass through zero and reach an applied field of about $+3$ Oe (vertical dashed line in Fig. 1), we arrive in a state where the thin Fe film rotates its magnetization direction and is thus coupled in a nonparallel configuration relative to the adjacent Ni layers. By further increasing the field, the magnetization of the Ni layer is also reversed and a parallel configuration is achieved once again. This behavior, while common in magnetic multilayers, has not been previously observed for magnetic elements in direct contact.

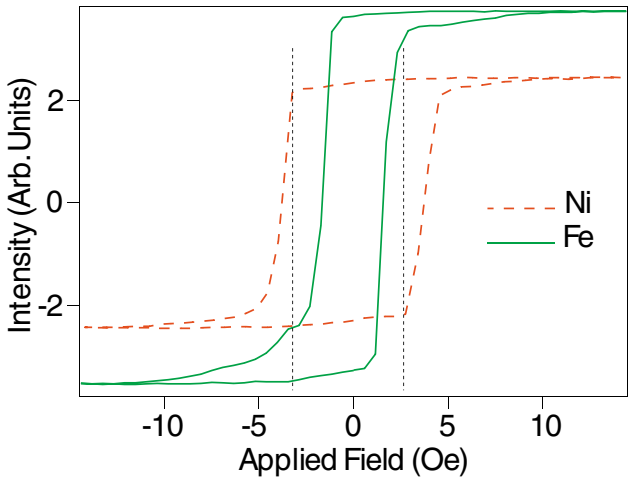

FIG. 1 (color online). The observed magnetic hysteresis data for the $Ni_2Fe_2Ni_8/Cu(001)$ trilayer system. The reflected intensity of circularly polarized soft x rays, with energies corresponding to the L_3 absorption edges of Fe (solid line) and Ni (dashed line), is measured as an applied magnetic field is cycled by means of an *in situ* solenoid. In this way we obtain elementally specific hysteresis loops.

While the coercive fields in Fig. 1 are small, they are consistent with previous thin film work [16]. We also note that the magnetization loops of Fig. 1 are similar to those reported for so-called *spring magnets* [17]. However, at 2 AL the thickness of the Fe component in the system presented here is sufficiently small as to rule out this interpretation.

There are three relevant contributions to the free energy: the magnetocrystalline anisotropy, the direct interlayer exchange energy, and the demagnetization energy (dipolar energy), the latter favoring an antiparallel alignment. From Fig. 1, and magnetic moments derived from XMCD data, we conclude that the energy required (the sum of demagnetization, anisotropy, and exchange energies) to obtain an antiparallel coupling between the Fe and Ni layers is of order 1 $\mu J/m^2$. From the magnetic moments [18], we also calculated the demagnetization energy from the expression for the classical magnetic dipole-dipole interaction adapted for the multilayer geometry [19] to be 0.3 $\mu J/m^2$. The in-plane anisotropy was also measured to be of this order of magnitude, implying that the strength of the direct interlayer exchange energy must be of order 1 $\mu J/m^2$ (~ 1 $\mu eV/atom$), i.e., 3–5 orders of magnitude smaller than that expected between $3d$ elements in direct contact.

Furthermore, the Fe and Ni anisotropy constants can be determined from our data using the product between the coercive field and the magnetization. One may imagine that the two Ni layers differ in coercivity and hence rotate independently. However, for both the Fe and the Ni components we obtain anisotropy constants of ~ 6 $\mu eV/atom$. While we cannot experimentally discriminate the top 2 AL from the bottom 8 AL of Ni, it appears logical that the upper and lower Ni layers revert at the same magnetic field. In none of the investigated samples did we find any evidence of such decoupling of the Ni layers.

To understand the microscopic mechanism behind our experimental findings we have performed first principles theoretical calculations of the exchange energy between the Fe and Ni atoms in this system. The calculations were made using the local spin density approximation for the geometry $Ni_2Fe_2Ni_8$ on a six layer Cu(001) slab using several theoretical techniques including a linear muffin-tin orbital (LMTO) method, within the atomic sphere approximation (ASA) Greens function method [20], a noncollinear LMTO-ASA [21] method, and a plane wave pseudopotential method [22]. Care was made in all computational steps, including basis set truncation, self-consistency criterion, and k-point summation. The ex-

change energies shown below were obtained via direct total energy calculations.

In order to investigate the influence of the effect of alloying and/or intermixing at the interfaces [23], we have calculated the direct interlayer exchange interaction as a function of intermixing. For simplicity this was not done for a surface geometry, but for a multilayer geometry, albeit with the same crystal structure (a multilayer in the 001 direction having an fcc crystal structure) and a lattice constant of fcc Cu. The geometry of this calculation is shown in Fig. 2, and our calculated values of the exchange energy as a function of the angle between Fe and Ni moments, Θ, are shown in Fig. 3. This calculation results in a ferromagnetic coupling between Fe and Ni atoms [24]. Interchanging every second Fe and Ni atom at the interface, to simulate intermixing, predicts antiferromagnetic or even noncollinear interactions. Indeed, Fig. 3 shows that a rotation of the Fe moments in a noncollinear fashion has a lower energy than if they are rotated collinearly. This is consistent with the behavior of bulk FeNi (Invar) alloys that form noncollinear magnetic structures for certain concentrations [25]. A mechanism that explains noncollinear magnetic exchange interactions has recently been suggested [26]. Two criteria were identified: that the Fermi level cuts through both the spin up and the spin down densities of state and that there is nesting between spin up and spin down states. For FeNi alloys, in the bulk as well as in the present system, the spin moments are not saturated and the first criterion of Lizarraga *et al.* [26] is indeed fulfilled. Since the nesting feature is more relevant for spin-spiral geometries, we have not pursued this analysis here. Again we emphasize that the data in Fig. 1 show that we have found a system where two ferromagnetic, monodomain, thin films in direct contact experience a vanishing interlayer exchange coupling.

Our theoretical calculations clearly indicate that the size of the interlayer exchange is very sensitive to small changes in the local structure (i.e., lattice constant and degree of alloying) of the interfaces and for certain geometries a vanishing value for the exchange interaction is found in accordance with the experimental observations presented here. However, a slight modification of the balance between the different energy contributions that determine the macroscopic magnetization—the exchange energy, the demagnetizing energy, and the contributions to the magnetocrystalline anisotropy energy (MAE) [27]—results in a drastically different magnetization behavior. For example, in a $Ni_2/Fe_{2.2}/Ni_{15}/Cu(001)$ sample the competition between the bulk and the surface contributions to the total MAE and a different interlayer exchange that could be attributed to a difference in the local geometry at the interfaces results in a multidomain magnetization behavior as witnessed by a severe rounding in the magnetization loops and the Fe and Ni layers having identical coercive fields.

In Fig. 4 we show a calculation of the direct interlayer exchange energy between Fe and Ni layers as a function of the lattice constant. It is clear that the calculated exchange energy is very small and crosses from positive to negative at a lattice constant that is close to what we expect in the Fe-Ni system presented here. In the past it has been shown that the nature of the exchange interaction in fcc Fe and in FeNi alloys does, indeed, depend critically on the interatomic distance [25] as described by the Bethe-Slater curve [28]. The results in Fig. 4 are consistent with these earlier works and is an important ingredient for explaining the observed magnetization curves (Fig. 1).

Since Fig. 4 suggests that the exchange interactions in this system are volume sensitive we also performed noncollinear calculations for somewhat smaller volumes. The results of these calculations show that, in the absence of interface alloying, the ferromagnetic state has a marginally lower energy than the antiferromagnetic state (in agreement with Fig. 4). However, when interface alloying is considered, the antiferromagnetic, or even noncollinear state, has a lower energy. In other words, the conclusions drawn from Figs. 2 and 3 do not change when one considers different lattice constants. Since a full structural characterization of the present system has not been performed, one can only speculate that the geometry of the interfaces is such that ferromagnetic and antiferromagnetic interactions cancel, yielding a vanishing direct interlayer ex-

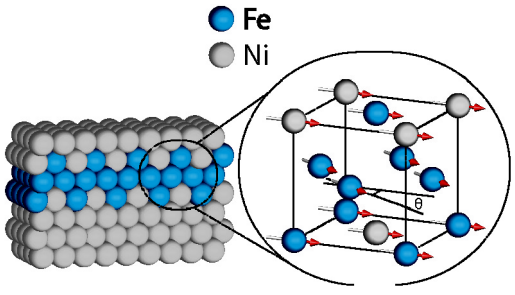

FIG. 2 (color online). An illustration of the geometry used in the theoretical calculations. The angle between the magnetic moments in the Fe and Ni layers is denoted Θ. In the noncollinear geometry half of the Fe magnetic moments rotate with Θ and the other half with $-\Theta$.

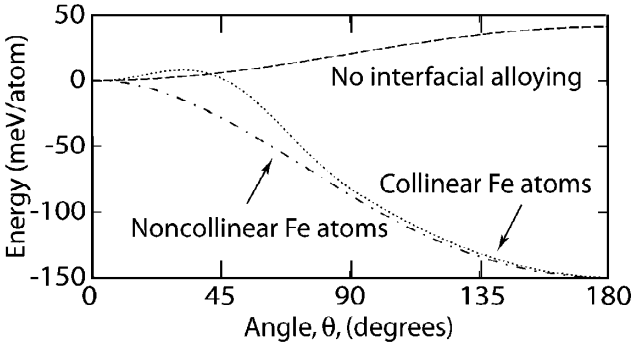

FIG. 3. The calculated exchange energy for the Fe_2/Ni_8 multilayer as a function of Θ, the angle between the magnetization direction of the Fe and Ni layers. (See Fig. 2.)

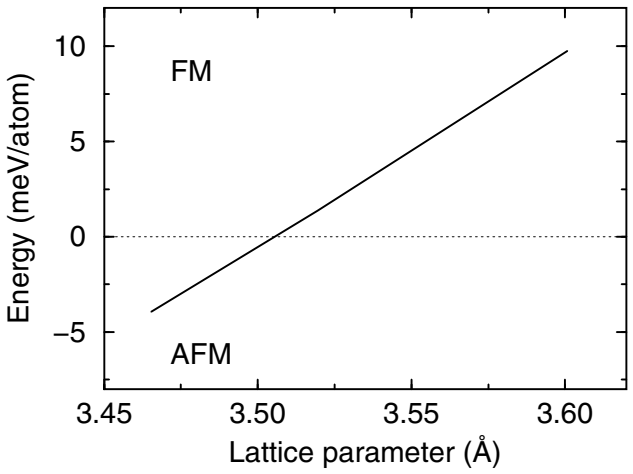

FIG. 4. The calculated energy difference between ferromagnetic and antiferromagnetic coupling of Fe and Ni trilayers as a function of the lattice constant.

change. It is clear from both our experimental findings and theoretical models that the necessary compensation occurs only in a very small thickness range.

To summarize, our discovery is the result of competing, ferromagnetic, antiferromagnetic, and noncollinear exchange interactions between 3*d* elements in direct contact that very delicately balance out to become vanishingly small. One may speculate that our discovery may also be found in layered structures that are composed of other elements exhibiting noncollinear couplings. Potential candidates exhibiting this behavior include bcc and fcc Mn, fcc Fe, and heavy rare-earth elements (e.g., Ho, Er, and Tm).

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