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Design of Co/Pd multilayer system with antiferromagnetic-to-ferromagnetic phase transition

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Among the known magnetic material systems, most are either purely antiferromagnetic or purely ferromagnetic at temperatures up to their critical temperature. There are only very few examples of materials that undergo a temperature dependent phase transition from an antiferromagnetic to a ferromagnetic phase or vice versa, and of these, only the chemically ordered alloy FeRh exhibits this transition near room temperature. Here we present a perpendicular anisotropy multilayer structure that by careful tuning of the constituent layers and their coupling mimics the magnetic behavior of FeRh, yet allows great flexibility in tuning its magnetic properties over a wide temperature range.

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The first order phase transition from an antiferromagnetic (AF) to a ferromagnetic (FM) phase in FeRh upon heating from room temperature to a transition temperature, T_{AF-FM} , of approximately 370 K was discovered in 1938 by Fallot.¹ In subsequent experiments using Mössbauer spectroscopy² and neutron diffraction³ the spin structure of the two phases was shown to be collinear with moments of $3.2\mu_B$ per Fe and $0.9\mu_B$ per Rh atom for the FM phase and $3.3\mu_B$ per Fe atom and no magnetic moment on the Rh for the AF phase. While other materials exhibiting similar first order AF-to-FM phase transitions such as Ru-doped CeFe₂ are known,⁴ FeRh is unique in that the phase transition occurs around room temperature. The transition temperature can be tuned over a wide range by substitutional doping: it increases with small additions of Ir or Pt, and decreases with small additions of Pd or Ni.⁵ Recently, FeRh was investigated with renewed interest for potential applications in thermally assisted magnetic recording and memory devices.⁶ However, one of the drawbacks of FeRh is the high deposition or postannealing temperature required to obtain the chemically ordered phase.

The aim of the present study is to design a multilayer structure that mimics the behavior of FeRh, utilizing the great flexibility offered by magnetic multilayers to tune all relevant magnetic properties.⁷ The basic idea is to use two layers with large perpendicular magnetic anisotropy and high Curie temperatures T_C separated by a layer providing AF coupling, and a layer with perpendicular anisotropy with a lower Curie temperature $T_{C,interlayer}$ in the range of the desired AF-FM transition temperature T_{AF-FM} . At room temperature, this system behaves as two antiferromagnetically coupled layers with a low net perpendicular magnetic moment in remanence. As the temperature is raised to approach $T_{C,interlayer}$, the magnetization of the interlayer is gradually reduced to zero, and consequently the coupling strength provided by the combination of this low- T_C layer and the adjacent AF coupling layer is reduced. Eventually, the effective coupling between the two high- K_U , high- T_C layers becomes

dominated by their dipolar fields, resulting in a parallel alignment of their moments at remanence and a net magnetic moment equal to the sum of the moments of the two high- T_C layers.

To prove the concept described above, we deposited sequences of Co/Pd and CoNi/Pd multilayers, as they allow great flexibility in adjusting the relevant parameters such as anisotropy, magnetization, Curie temperature, and intralayer as well as interlayer exchange coupling. The basic structure, as shown in Fig. 1, consists of two Co/Pd multilayers with large perpendicular magnetic anisotropy and high T_C separated by a Co/Ru (7 Å)/Co structure providing AF coupling and a CoNi/Pd multilayer with perpendicular anisotropy and $T_{C,interlayer}$ in the range of the desired AF-FM transition temperature T_{AF-FM} . The multilayer samples were deposited on glass substrates by magnetron sputtering from element targets in a commercial sputter system. An Ar gas pressure of 1.4 mTorr was used for sputtering all layers except the topmost Co/Pd multilayer stack. This last multilayer was deposited at 5 mTorr in order to achieve a somewhat larger coercivity compared to the previous layers. The deposition rates of all layers were in the range of 0.5–1 Å/s. Typical film thicknesses and compositions of the constituting multilayers were [Co (5 Å)/Pd (7.5 Å)] for the first high- T_C multilayer, Co (5 Å)/Ru (7 Å)/Co (5 Å) for the AF coupling layer, [Co₅₀Ni₅₀ (2.5 Å)/Pd (12 Å)] for the separation layer and [Co (3.5 Å)/Pd (7.5 Å)] for the second high- T_C multilayer.

FIG. 1. (Color online) Sample structure showing the four basic building blocks: two Co/Pd multilayers with large perpendicular magnetic anisotropy and a high Curie temperature T_C , a Co/Ru (7 Å)/Co structure providing AF coupling, and a CoNi/Pd multilayer with perpendicular anisotropy and $T_{C,interlayer}$ in the range of the desired AF-FM transition temperature T_{AF-FM} .

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FIG. 2. Temperature dependence of the remanent magnetization M_R and the saturation magnetization M_S of a $[\text{Co}_{50}\text{Ni}_{50} (2.5 \text{ Å})/\text{Pd} (12 \text{ Å})] \times 12$ multilayer. T_{sro} denotes the temperature at which the effective anisotropy of the film changes from perpendicular to in-plane orientation $T_{\text{C, interlayer}}$ denotes the Curie temperature.

Thin seed and cap layers of about 50 and 25 Å Pd were used. Film thicknesses were measured on calibration samples by grazing angle x-ray diffraction, and the composition of the CoNi alloy used for the low- T_C layer was verified using Rutherford backscattering. Hysteresis loops as well as temperature scans of all structures and the saturation magnetization M_S and anisotropy constant K_1 of the constituent multilayers were measured in a maximum field of 2 T using a commercial vibrating sample magnetometer.

The two Co/Pd multilayers have an estimated T_C of about 400–500 °C (it is actually difficult to measure T_C experimentally, as at temperatures exceeding about 300 °C the individual layers of the multilayer start interdiffusing).⁸ In contrast, the CoNi/Pd multilayer was tuned to obtain a $T_{\text{C, interlayer}}$ of approximately 100 °C. Due to the more rapid decrease with temperature of the anisotropy relative to the magnetization, the CoNi/Pd multilayer shows a spin reorientation from perpendicular to in-plane at a temperature T_{sro} of about 40 °C, as shown in Fig. 2. At room temperature, the complete multilayer system behaves as two antiferromagnetically coupled multilayers, resulting in two shifted hysteresis loops with a low net perpendicular magnetic moment in remanence. By proper choice of film thicknesses, compositions, and multilayer repeats the net moment at room temperature can be adjusted to zero, as shown in Fig. 3(a) for a $[\text{Co}/\text{Pd}] \times 5/\text{Co}/\text{Ru}/\text{Co}/[\text{CoNi}/\text{Pd}] \times 3/[\text{Co}/\text{Pd}] \times 3$ sample. As the temperature of the sample is raised, at T_{sro} first the effective perpendicular anisotropy of the CoNi/Pd multilayer and subsequently at $T_{\text{C, interlayer}}$ its magnetization are reduced to zero. Without the mediation of the CoNi/Pd multilayer, the coupling strength provided by the Co/Ru/Co layers is also greatly reduced and the effective coupling between the two Co/Pd multilayers becomes dominated by their dipolar fields, resulting in a parallel orientation. Figure 3(b) shows the temperature dependence of the remanent magnetization, measured after applying a perpendicular field of 1 kG at each temperature.

Interestingly, while T_C of CoNi/Pd multilayers generally does not vary with the number of repeats N , $T_{\text{AF-FM}}$ of the complete structure increases significantly with decreasing number of CoNi/Pd repeats, as shown in Fig. 4, for a series of $[\text{Co}/\text{Pd}] \times 5/\text{Co}/\text{Ru}/\text{Co}/[\text{CoNi}/\text{Pd}] \times N/[\text{Co}/\text{Pd}] \times 3$ samples. This reflects the influence of the adjacent Co and Co/Pd layers on the band structure and the resulting magnetic properties of the CoNi/Pd multilayer as

FIG. 3. (a) VSM hysteresis loops of a $[\text{Co}/\text{Pd}] \times 5/\text{Co}/\text{Ru}/\text{Co}/[\text{CoNi}/\text{Pd}] \times 3/[\text{Co}/\text{Pd}] \times 3$ sample measured at 25 °C (line) and 125 °C (symbols). (b) Remanent moment of that sample as a function of temperature, measured after applying a field of 1 kG at each temperature.

described by Wang and Mills,⁹ and seen experimentally in studies of $\text{FeF}_2/\text{CoF}_2$ superlattices reported by Ramos *et al.*¹⁰ Here a decay length of the same order as the exchange length of the material equivalent to approximately four repetitions, i.e., $l_d = 6 \text{ nm}$, can be estimated for this specific case from the data in Fig. 4. Moreover, for CoNi/Pd interlayers thicker than the decay length, $T_{\text{AF-FM}}$ converges to a value below $T_{\text{C, interlayer}}$ and slightly above T_{sro} , indicating that the spin reorientation is sufficient to trigger the AF-FM transition.

Finally, in the following, we discuss the similarities and differences between FeRh and the multilayer system presented here. Recently a unified theoretical description of FeRh and related alloys based on first-principles calculations for the exchange fields of the Fe atomic moment and the induced atomic moments on Rh was proposed.¹¹ According to these calculations in both the AF and FM phases, the governing magnetic energy is the exchange energy, specifi-

FIG. 4. AF-to-FM coupling transition temperature $T_{\text{AF-FM}}$, as a function of the number of repeats N in the CoNi/Pd multilayer in a series of $[\text{Co}/\text{Pd}] \times 5/\text{Co}/\text{Ru}/\text{Co}/[\text{CoNi}/\text{Pd}] \times N/[\text{Co}/\text{Pd}] \times 3$.

cally the moment induced by the Fe on the Rh, and its interplay with the lattice spacing and the resulting band structure. In contrast, in the multilayer system presented here in the AF phase, it is the significantly weaker Ruderman–Kittel–Kasuya–Yoshida (RKKY) interaction across the Co/Ru/Co coupling layer that results in the AF coupling and the zero net moment, while in the high temperature FM phase it is the dipolar fields of the two high T_C multilayers that cause a parallel alignment. Consequently the energy scales governing the magnetic properties of the two materials are vastly different. At temperatures close to the AF-FM transition in FeRh, the transition can be induced by an external magnetic field, and the transition shifts linearly by about 8–9 K/T,¹² i.e., for a sample with T_{AF-FM} of approx 100 °C a magnetic field of the order of 8 T is required to induce the phase transition at room temperature. In contrast, as shown in Fig. 3, in the multilayer system presented here, only 0.18 T are required to overcome the RKKY coupling across the Co/Ru/Co coupling layer. Similarly, in the FM phase, FeRh is still governed by the exchange interaction, so the internal fields associated with, e.g., crystal defects are very large and accordingly in experimental samples no external fields are required to induce the AF-FM transition. In full films in a single-domain state dipolar fields are close to zero, so that in the multilayer system here small external fields are still required to induce the AF-FM transition even at temperatures above T_{AF-FM} . However, external fields are not required in samples in a multidomain state¹³ or nanostructured samples, such as, e.g., patterned media, one of the envisioned applications of such multilayer systems.

In summary, we have demonstrated a multilayer system that with increasing temperature shows a transition from antiparallel alignment with zero net remanent magnetic mo-

ment to parallel alignment with a large net remanent magnetic moment. This is achieved by coupling two Co/Pd multilayers via a Co/Ru (7 Å)/Co structure providing AF coupling and a CoNi/Pd multilayer with perpendicular anisotropy and tunable T_C in the range of the desired AF-FM transition temperature. In terms of the temperature dependence of the remanent magnetization, this behavior mimics that of the natural superlattice FeRh, which undergoes a first order phase transition from an AF to a FM phase.

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