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Structure and magnetic properties of exchange-biased polycrystalline Fe/MnPd bilayers

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We report the fabrication of polycrystalline Fe/MnPd bilayers, which show the exchange-bias effects. We show that the exchange-bias effect, described by unidirectional anisotropy, is also accompanied by induced uniaxial and fourfold in-plane anisotropy contributions in the system. Using the Stoner-Wohlfarth model, all in-plane anisotropy contributions to the total free energy are determined for the polycrystalline samples from the fit of the in-plane angular dependence of H_{eb} and the coercivity H_c. A mechanism explaining the presence of the induced higher-order anisotropy contributions in an exchange-bias system is proposed.

I. Introduction. Exchange coupling at the interface between a ferromagnetic (FM) and an antiferromagnetic (AF) layer can cause exchange biasing, i.e., a shift of the hysteresis loop of the FM layer along the field axis. This effect is characterized by an exchange-bias field H_{eb}, which is often described as a result of an acting in-plane unidirectional anisotropy.[1] Exchange biasing has attracted wide interest because of its technological use in magnetic sensors and high density magnetic recording systems.

Although the exchange-bias phenomenon has been experimentally studied to some extent, the physics of FM/AF interface exchange coupling still remains a subject of discussion.[2–7] Earlier theories predict exchange-bias fields of orders of magnitude larger than those observed.[1] Mauri *et al.* provided an explanation for the reduced exchange-bias fields by showing that the formation of a domain wall parallel to the interface in the AF layer can greatly reduce the energy required to reverse the magnetization.[2] Malozemoff interpreted the exchange-bias effect in terms of random exchange fields due to interface roughness.[3] Both the theories of Mauri and Malozemoff predict correct order-of-magnitude results for H_{eb}. It was recently theoretically shown[4,8] that a spin-flop reorientation in the AF layer can also lead to the exchange-bias effect. The latter models imply a perpendicular orientation between the magnetizations of the FM and the AF layer, as it was found in $\mathrm{Fe_3O_4/CoO}$ (Ref. 5) and $\mathrm{Fe/FeF_2}$.[6]

The probably most used exchange-biased system contains $\mathrm{Fe_{50}Mn_{50}}$ as the antiferromagnetic layer, which, however, has a relatively low Néel temperature (∼200 °C) and is easily corroding. Many efforts have been made to search for a suitable pinning layer in antiferromagnetic sensors and recording heads. In this context MnPd is a very promising material. MnPd belongs to the same group of alloys as $\mathrm{Mn_{1-x}Pt_x}$, which was already used for exchange biasing.[9,10] MnPd has a CuAu-I-type ordered face-centered-tetragonal (fct) structure with lattice constants $a = 4.07$ Å and $c = 3.58$ Å. Previous studies[11] revealed that the ordered fct structure of MnPd possesses an antiferromagnetic phase with a Néel temperature as high as 540 °C. The direction of the Mn magnetic moment is believed to be aligned along either the ⟨100⟩ or the ⟨110⟩ directions, which in any case results in a compensated spin arrangement of the MnPd (001) plane.[11] However, to our knowledge, MnPd was practically not studied as a potential exchange-bias layer.

In this paper we report a detailed study of the structure and the magnetic properties of polycrystalline Fe/MnPd exchange-bias systems. We show that the exchange-bias effect, described by an unidirectional anisotropy, is also accompanied by induced uniaxial and fourfold in-plane anisotropy contributions in the system. Using the Stoner-Wohlfarth model, all in-plane anisotropy contributions to the total free energy are determined for the polycrystalline samples from the fit of the in-plane angular dependence of H_{eb} and the coercivity H_c.

II. Experimental. The samples were grown in an UHV evaporation system with a base pressure better than 5×10^{-9} mbar. The polycrystalline samples were grown at room temperature (RT) on thermally oxidized Si substrates with the structure $\mathrm{Si/SiO_2/Fe}(t_F)/\mathrm{MnPd}(t_{\mathrm{AF}})/\mathrm{Pd}(20\,\text{Å})$ ($t_{\mathrm{AF}} = 200$ Å; $t_F = 30$Å, 50 Å) or a wedge-shaped Fe layer with thickness t_F varying from 20 to 160 Å, respectively). Fe and Pd were evaporated by means of a multipocket *e*-beam evaporator, while Mn was evaporated from an effusion cell. The growth rates were carefully controlled by a quartz microbalance and were 0.1 Å/s for Fe and Pd and 0.2 Å/s for MnPd, respectively. In all cases a 20-Å-thick Pd cap layer was used for protecting the sample against oxidization. A magnetic field of 50 Oe was applied in the sample plane during growth. The chemical analyses of the prepared films were performed *in situ* by means of a calibrated Auger-electron spectrometer. Additional structural information was obtained by *ex situ* x-ray diffraction. The static magnetic properties of the prepared films were measured at room temperature by the magneto-optic Kerr effect (MOKE) magnetometry.

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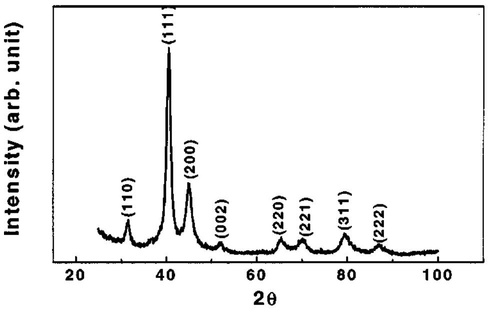

FIG. 1. X-ray diffraction scan of the polycrystalline sample Si/SiO$_2$/Fe(30 Å)/MnPd(200 Å)/Pd(20 Å).

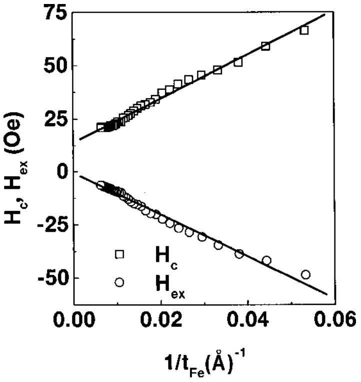

FIG. 2. The exchange-bias field H_{eb} and the coercive field H_c of the polycrystalline samples Si/SiO$_2$/Fe(t_{Fe})/MnPd(200 Å)/Pd(20 Å) versus $1/t_{\mathrm{Fe}}$.

III. Results and discussion. For MnNi, MnPt, and Pd-Pt-Mn films, which have the same CuAu-I-type structure as MnPd, it was previously reported that as-deposited films mainly contain the nonmagnetic disordered fcc phase with almost no indication of the AF fct phase.[9,10] An additional annealing step or an elevated growth temperature was found to be necessary for obtaining the antiferromagnetic phase. However, under these circumstances an interfacial diffusion is almost inevitable. In our study, we found that by using an appropriate underneath layer, as-deposited AF fct MnPd films can be obtained at room temperature. Figure 1 illustrates the results of the x-ray diffraction analysis of a Si/SiO$_2$/Fe(30 Å)/MnPd(200 Å)/Pd(20 Å) sample. All observed peaks can be well assigned assuming the fct phase of MnPd. A splitting between the (200) and (002) peaks, also indicating the presence of the fct phase, is clearly seen in Fig. 1. The corresponding lattice constants calculated from the positions of the (111) and (200) peaks ($a=4.02$ Å, $c=3.56$ Å) are consistent with those obtained earlier for the ordered fct AF phase of bulk MnPd.[11]

Polycrystalline Fe films, grown on Si/SiO$_2$, were found to exhibit no observable magnetic in-plane anisotropy. This is most likely due to an averaging of the intrinsic anisotropies of bcc Fe over a large amount of small grains. On the other hand, the Fe/MnPd structures show a unidirectional anisotropy causing the effect of the exchange bias. The directions of the exchange-bias field in all investigated samples were found to coincide with the direction of the field applied during the sample deposition. The thickness dependencies of the exchange-bias field H_{eb} and the coercive field H_c for the Si/SiO$_2$/Fe(20–160 Å)/MnPd(200 Å)/Pd(20 Å) sample with a wedge-shaped Fe film are presented in Fig. 2. The exchange-bias field varies from about 50 to 5 Oe, while the coercive field varies from about 65 to 20 Oe with increasing Fe thickness. As expected, a linear dependence of both the exchange-bias field H_{eb} and the coercive field H_c on the inverse Fe layer thickness indicates the interfacial nature of the effect.

The linear dependence of the exchange-bias field H_{eb} was also found in many experiments and explained by previous studies.[2,3] Supposing that there is a domain wall formed in the antiferromagnetic layer in FM/AF bilayers, Mauri[2] shows that the exchange-bias field H_{eb} can be written as $H_{\mathrm{eb}}=2(AK)^{1/2}/t_{\mathrm{FM}}/M_{\mathrm{FM}}$, where t_{FM} and M_{FM} are the thickness and the saturation magnetization of the FM layer, respectively; $2(AK)^{1/2}$ is the domain-wall energy in the antiferromagnetic layer, where A and K are the exchange stiffness and the crystalline anisotropy in the antiferromagnet, respectively. The idea of the formation of a domain wall in an antiferromagnet is further introduced by Koon[4] and Stiles and McMichael[12] to explain the high field rotational hysteresis and other properties found in FM/AF exchange-bias bilayers. The interfacial roughness can also lead to the linear dependence of the exchange-bias field H_{eb} on the inverse FM layer thickness, as provided by Malozemoff,[3] who suggested that the random fields at the interface will cause the antiferromagnet to break up into domains when the system is cooled through the ordering temperature. The size of the domains is inversely proportional to the exchange-bias field. The interfacial roughness effect on the exchange-bias field was also confirmed by a recent micromagnetics calculation.[8]

However, the study on the coercivity in FM/AF bilayers was almost ignored previously, although an enhancement of the coercivity in exchange-bias bilayers was generally found. Only recently has it been shown that the spin-flop exchange coupling at the FM/AF interfaces can induce uniaxial anisotropy, which could account for the enhanced coercivity.[8] Our recent study[13] on the coercivity of Fe/MnPd bilayers, which is based on Mauri's calculation, also reveals that the linear dependence of the enhanced coercivity on the inverse FM layer thickness, as shown in Fig. 2, could be ascribed to the higher-order anisotropies induced by the exchange coupling in FM/AF bilayers.

To compare our results with those of other frequently used exchange-bias systems, the magnitude of the exchange-bias effect can be described in terms of an interfacial energy per unit area ΔE, which is independent of the ferromagnetic material and its thickness:[14] $\Delta E=M_{\mathrm{FM}}t_{\mathrm{FM}}H_{\mathrm{eb}}$. The obtained interfacial energy value for the system with MnPd is about 0.017 erg/cm^2, using $M_{\mathrm{FM}}=1700\,\mathrm{emu/cm}^3$ for Fe. We note that this value is slightly smaller than those for the as-deposited structures with FeMn, which are typically from 0.02 to 0.2 erg/cm^2.[14] However, it is much larger than those for the as-deposited system with NiMn, which is about 0.002 erg/cm^2.[14] It is also comparable with those annealed structures with MnPt, which have interfacial energy normally ranging from 0.02 to 0.32 erg/cm^2.[14] These results show that MnPd is a promising antiferromagnetic material that can be used in the FM/AF exchange-bias system.

It is also interesting to note that the in-plane unidirectional anisotropy corresponding to the exchange bias is ac-

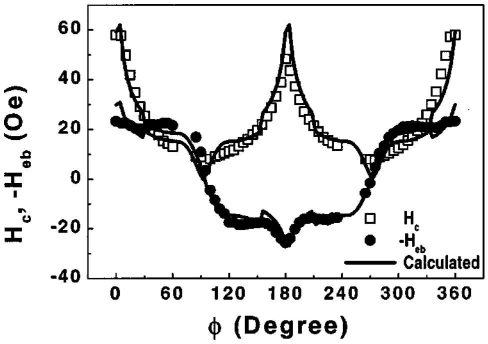

FIG. 3. The angular dependence of the exchange-bias field H_{eb} and the coercivive field H_c for the sample Si/SiO$_2$/Fe(50 Å)/MnPd(200 Å)/Pd(20 Å). The solid lines show the result of the calculations using the free-energy expression Eq. (1).

companied by a uniaxial and a fourfold anisotropy. This is demonstrated by the dependencies of H_c and H_{eb} on the in-plane angle θ of the applied external field, presented in Fig. 3 for the Si/SiO$_2$/Fe(50 Å)/MnPd(200 Å)/Pd(20 Å) sample. The maximum values of H_c and H_{eb} occur at $\theta=0$ and 180°, while the minimum values of H_c and H_{eb} occur at $\theta=90°$ and 270°. It is also obviously seen in Fig. 3 that the angular is not a simple cos(θ) behavior, as it was found in polycrystalline NiFe/FeMn samples under almost identical conditions.[15] Note that a similar complex in-plane angular dependence of H_{eb} was obtained for an epitaxial NiFe/CoO bilayer.[7]

Although the measurement of angular dependence of the exchange-bias field and the coercivity has an important role in understanding the nature of the exchange-bias effect since the exchange-bias effect may be strongly affected by the various directions of the spins at the interface of different layers, there have been only a few such studies until now.[7,15–17] While it was found that the polycrystalline NiFe/FeMn samples presented a pure cos(θ) dependence of the exchange-bias field, no higher-order symmetry was found;[15] the epitaxial samples exhibited a large higher-order symmetry due to exchange coupling.[17] This might be due to the relatively large induced anisotropies in epitaxial system as discussed below. A behavior that is quite similar to our results and that also clearly indicates the existence of a higher-order symmetry of the exchange-bias field was also found for the epitaxial NiFe/CoO system.[7] We note that the use of Fe other than NiFe as the ferromagnet in the Fe/MnPd system, which may cause larger induced both uniaxial and fourfold anisotropies, leads to a more complex behavior of the angular dependence of the exchange-bias field as revealed by our studies. By taking a pure symmetric consideration, Ambrose *et al.*[7] also tried to describe the angular dependence of the exchange-bias field for the NiFe/CoO system with $H_{\mathrm{eb}}(\theta)=\sum_{n=\mathrm{odd}} b_n \cos n\theta$, where b_n is a constant. According to this, they proposed that the largest exchange-bias field should not occur at $\theta=0°$ and 180° but at 135° and 225° as they found for the NiFe/CoO system, although this is not observed in our results and other experiments.[15,16]

Another interesting thing is that the coercivity of all the systems shows a sharp peak around $\theta=0$ and 180°, i.e., the easy direction of the magnetization, while it is relatively smaller and flat at other angles. Although the enhancement of coercivity in the FM/AF system compared with the pure FM layer is quite evident, only just recently has more attention been given to the study of the coercivity and the magnetization process.[7,8,16,18,19] Ambrose *et al.*[7] pointed out that the coercivity can only be realized when even terms of the $\cos n\theta$ are included in the free energy of the system, which means higher-order anisotropies may be the origin of the coercivity for the FM/AF system. Based on a model, which is an extension of the random-field theory,[3] Dimitrov *et al.*[16] recently proposed that the pinning of the domain wall in the FM layer by the local energy minima of the AF random exchange field can enhance the coercivity more than two orders of magnitude. More important, they prospected that the magnetization reversal is by domain-wall nucleation and propagation when the field is applied at a hard direction of the magnetization while the reversal is complicated by a mixture of partial rotation and domain-wall propagation at other angles, which causes a sharp increase of the coercivity around 0° and 180°. Similar results were also obtained by other studies.[19] Other models that take interface roughness effect into consideration have also been developed to describe the enhanced coercivity.[18] Due to the large contributions of higher-order anisotropies in our system, it is reasonable to believe that the enhanced coercivity in our system is mainly produced by these contributions.[13]

The measured in-plane angular dependencies of H_c and H_{eb} can be described by simulating the remagnetization process using the Stoner-Wohlfarth model with a coherent rotation of the magnetization assuming a free-energy expression:[17]

$$F_{\mathrm{ani}}=K_p^{(1)}\cos(\phi)+K_p^{(2)}\cos^2(\phi-\phi_{\mathrm{off}}^{(2)})+K_p^{(4)}\cos^2 \times(\phi-\phi_{\mathrm{off}}^{(4)})\sin^2(\phi-\phi_{\mathrm{off}}^{(4)})-HM_S\cos(\theta-\phi), \qquad (1)$$

where $K_p^{(1)}$, $K_p^{(2)}$, and $K_p^{(4)}$ are the unidirectional, uniaxial, and fourfold anisotropy constants, respectively; ϕ is the in-plane angle of the magnetization with respect to the easy direction of $K_p^{(1)}$; $\phi_{\mathrm{off}}^{(2)}$ and $\phi_{\mathrm{off}}^{(4)}$ are the angular offsets between the easy direction of $K_p^{(1)}$ and $K_p^{(2)}$ and $K_p^{(4)}$, respectively; and θ is the in-plane angle of the applied magnetic field H, and M_s is the magnetization. Note here that in addition a third-order term in Eq. (1), which is normally forbidden from the symmetry considerations, is allowed in systems with the exchange-bias effect. The experiment, however, shows that in the investigated samples this term, as well as a sixfold anisotropy contribution, is negligible. It also shows that $K_p^{(2)}<0$, $K_p^{(4)}>0$, and $\phi_{\mathrm{off}}^{(2)}=\phi_{\mathrm{off}}^{(4)}=0$, i.e., the easy direction of $K_p^{(1)}$ coincides with an easy axis of both the uniaxial and the fourfold anisotropy. Figure 3 also shows the result of the fitting procedure by solid lines. We find a very good agreement between the experiment and the simulation.

The unidirectional, uniaxial, and fourfold anisotropy constants of the wedge-shaped sample Si/SiO$_2$/Fe(t_F)/MnPd(200 Å)/Pd(20 Å) with the Fe thickness t_F varying from 20 Å to 160 Å obtained from the fit are plotted in Fig. 4. as functions of the Fe thickness. Both induced in-plane anisotropy contributions $K_p^{(2)}$ and $K_p^{(4)}$ are comparable in strength with $K_p^{(1)}$. Moreover, $K_p^{(2)}$ and $K_p^{(4)}$ decrease with increasing Fe thickness, obeying a $1/d_{\mathrm{Fe}}$-scale law in a similar way as $K_p^{(1)}$ does, indicating their interfacial origin.

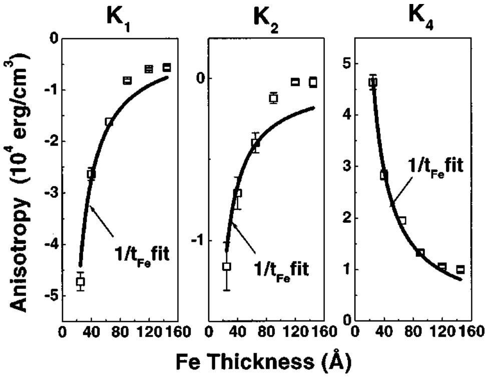

FIG. 4. The unidirectional, uniaxial, and fourfold anisotropy constants, $K_p^{(1)}$, $K_p^{(2)}$, and $K_p^{(4)}$, respectively, obtained from the simultaneous fit of $H_{\mathrm{eb}}(\phi)$ and $H_c(\phi)$ and plotted versus $1/t_{\mathrm{Fe}}$.

The induced higher-order anisotropy contributions caused by FM-AF exchange coupling were also found by other studies. By using the Brillouin light scattering (BLS) method Mathiea *et al.*[20] found that in an epitaxial NiFe/FeMn system, the uniaxial and fourfold anisotropy contributions are largely modified by an amount that also scales with the NiFe thickness. In NiFe/NiCoO (Ref. 16) and NiFe/CoO (Ref. 7) systems, since it is quite clear that the anisotropy contributions for the pure NiFe layer are quite smaller than those required to produce the hysteresis loops for the system, the induced uniaxial and fourfold anisotropy contributions were also ascribed to the interfacial interaction of the FM and AF layer, which produces the unidirectional anisotropy. Obviously, the observed large higher-order anisotropy contributions in the FM/AF system cannot only be ascribed to the intrinsic magnetocrystalline of the FM or AF layer. From the theoretical point of view the interplay between the exchange-bias effect and the induced anisotropy contributions is not clearly understood yet, although attempts to consider this problem theoretically have been made recently.[8] However, no detailed model has been set up so far to explain the induced higher-order anisotropy contributions accompanying the exchange-biasing effect.

Recent studies found that FM spins tend to align perpendicular to the AF easy axis, which may account mainly for the induced higher-order anisotropies.[4,5] The perpendicular coupling can be formed due to the spin frustration at the interfaces, in which the FM spins minimize the energy when they align perpendicular to the AF easy axis. Such effective exchange coupling is shown related to the ''spin-flop'' state in the antiferromagnet, which although it does not lead to the exchange coupling, it does introduce a uniaxial anisotropy which leads to an enhanced coercivity.[8] For the polycrystalline Fe layer, the total magnetocrystalline anisotropy tends to be zero due to the average of the local cubic anisotropy over all the Fe grains. Thus for the polycrystalline Fe/MnPd bilayers, the induced uniaxial and cubic anisotropies cannot be due to the intrinsic properties of the Fe layer. Considering the exchange coupling between the Fe spins and those AF spins from different sublattices of the MnPd layer, which may have different orientations at the interfaces, it is reasonable to believe that the higher-order anisotropies found in our study may be caused by these nonlinear couplings of FM and AF spins. On the other hand, although the average anisotropy of the polycrystalline Fe layer is almost zero, the local cubic anisotropies in fine grains may also be a benefit to such a nonlinear FM/AF exchange coupling, which may lead to a relatively strong induced higher-order anisotropies in the Fe/MnPd bilayer. As a matter of fact, since both the uniaxial and fourfold anisotropies are due to the exchange interaction at the interface between the FM and AF layers, they should have the same interfacial character as the unidirectional anisotropy, typically the linear dependence on the inverse Fe thickness, which is in fact revealed by our study.

IV. Conclusions. To summarize, we have reported studies of an exchange-bias system Fe/MnPd. We show that the exchange-bias effect, described by a unidirectional anisotropy, is also accompanied by induced uniaxial and fourfold in-plane anisotropy contributions in the system. From fitting the angular dependence of the exchange-bias field H_{eb} and the coercive field H_c, all induced in-plane anisotropy contributions to the total free energy of the polycrystalline samples are determined. A mechanism explaining the presence of the induced higher-order anisotropy contributions in a polycrystalline exchange-bias system is proposed.

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