Disorder, ordering, and superstructures of (FePt) and (CoPt) on Pt substrates

L. Szunyogh

*Department of Theoretical Physics, Technical University Budapest, Budafoki út. 8, H-1521 Budapest, Hungary
and Center of Computational Materials Science, Technical University Vienna, Getreidemarkt 9/158, A-1060 Wien, Austria*

P. Weinberger

*Institut für Technische Elektrochemie und Festkörperchemie and Center of Computational Materials Science,
Technical University Vienna, Getreidemarkt 9/158, A-1060 Wien, Austria*

C. Sommers

Laboratoire de Physique des Solides, Batiment 510, Campus d'Orsay, 91405 Orsay, France
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The magnetic anisotropy energy of interdiffused superstructures of (FePt) and of (CoPt) on Pt(100) and Pt(111) is calculated in terms of the spin-polarized relativistic screened Korringa-Kohn-Rostoker method. It is shown that interdiffusion between Pt and the magnetic $3d$ metal or, oppositely, ordering into a superstructure, very strongly influences the size of the perpendicular magnetism to be expected. [S0163-1829(99)02038-X]

In recent years (Fe,Co)/(Pt,Pd) multilayer systems[1] have attracted much attention in relation to magnetic data-storage applications. In particular Co/Pt-type superstructures[2–5] as well as the equivalent Fe/Pt systems[6–8] were investigated intensively using various experimental techniques. Most studies were directed to investigations of the large perpendicular magnetism inherent to these systems. In a recent paper[9] we studied the magnetic properties of thin films of Co and of (CoPt) superstructures on Pt(100) and Pt(111), leaving out the delicate question of interdiffusion at the Co/Pt interfaces. It was found that irrespective of the number of repetitions in the case of (CoPt) superstructures on both types of substrates the magnetization is strongly orientated perpendicular to the surface, whereas for not too thin films of Co on these substrates the orientation of the magnetization is in plane. In the present paper first attempts are made to describe the magnetic anisotropy energy on an *ab initio* level, by taking into account effects of ordering into superstructures. For this purpose we applied the relativistic spin-polarized screened Korringa-Kohn-Rostoker method[10–12] within the framework of the coherent potential approximation (CPA) for layered systems[13] to the set of systems shown in Table I. In there c_d is used as a shorthand notation for the degree of interdiffusion: $c_d = 0.5$ implies a homogeneously (statistically) disordered equiconcentrational alloy, $c_d = 1.0$ an ordered superstructure. The interdiffusion parameter c_d is simply defined as the $3d$-metal concentration in atomic layers with odd layer indices. If r denotes the number of repetitions of, e.g., the unit CoPt in a given superstructure, then the total number of atomic layers is $2r$. Consequently an odd number of alloyed overlayers leads to a $3d$-metal terminated superstructure, an even number to a Pt terminated superstructure. In order to show both cases of termination, effects of repetition are discussed for an even number of atomic layers and ordering phenomena for an odd number of (alloyed) atomic layers.

The magnetic anisotropy energy ΔE_a,

$$\Delta E_a = E(\|) - E(\perp), \tag{1}$$

is defined as the energy difference between a uniform in-plane (perpendicular to the surface normal in all planes of atoms) and a uniform perpendicular (along the surface nor-

TABLE I. Multilayer systems.

"Interdiffusion"	$\longrightarrow$	"Superstructure" M terminated	"Superstructure" Pt terminated
$Pt(hkl)/(M_{1-c_d}Pt_{c_d})_n$		$Pt(hkl)/(M_{1-c_d}Pt_{c_d})_n$	$Pt(hkl)/(Fe/Pt)_r$

$$r = (n-1)/2 \qquad r = (n-1)/2 \qquad r = 1, 2, \ldots, 15$$
$$n = 15 \qquad n = 15 \qquad n = 2r$$
$$0.5 \le c_d \le 1.0 \qquad c_d = 1.0$$
$$M = Fe, Co \qquad M = Fe, Co$$
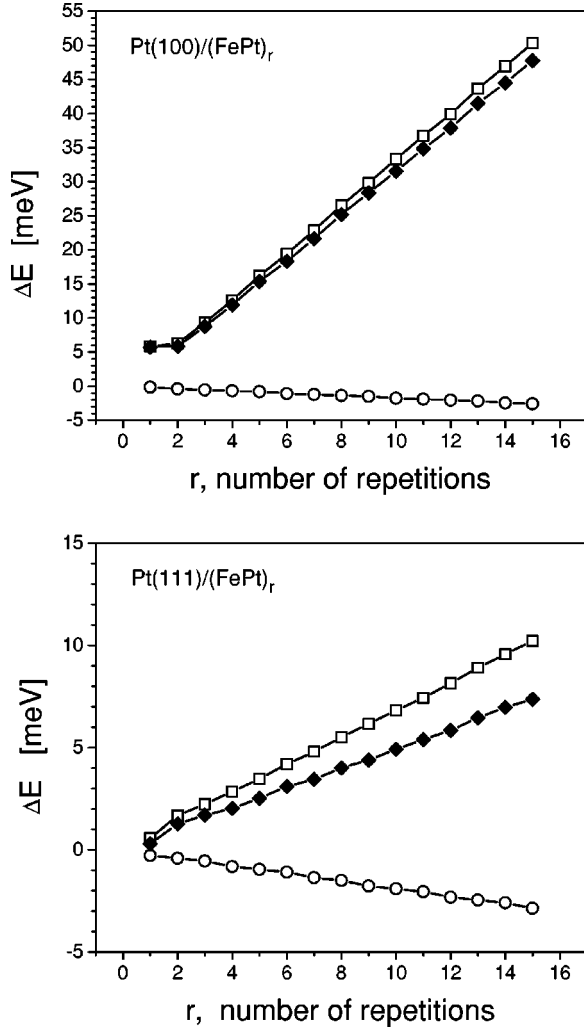
$$(hkl) = (100), (111)$$

FIG. 1. Band energy (open squares) and magnetic dipole-dipole interaction (open circles) contribution to the magnetic anisotropy energy (diamonds) of superstructures of (FePt) on Pt(100) (top) and Pt(111) (bottom) as a function of repetition.

mal in all planes of atoms) orientation of the magnetization of the system. In terms of the force theorem approximation,[10,12] ΔE_a is given by the sum of the respective band energy difference ΔE_b and the magnetic dipole-dipole energy contribution ΔE_{dd},

$$\Delta E_a = \Delta E_b + \Delta E_{dd}. \tag{2}$$

In the case of an inhomogeneously alloyed system, i.e., by using an inhomogeneous CPA condition,[13,14] ΔE_b arises from concentration weighted component- and layer-resolved band energy differences $\Delta E_{b,p}^\alpha$,

$$\Delta E_b = \sum_{p=1}^{n} \sum_{\alpha=A,B} c_p^\alpha \Delta E_{b,p}^\alpha, \tag{3}$$

where c_p^α denotes the concentration of component α in the pth atomic layer and n is the total number of atomic layers considered, including, e.g., vacuum layers. In a similar manner ΔE_{dd} refers to averaged magnetic moments.[14] A description of the computational details can be found in Ref. 9.

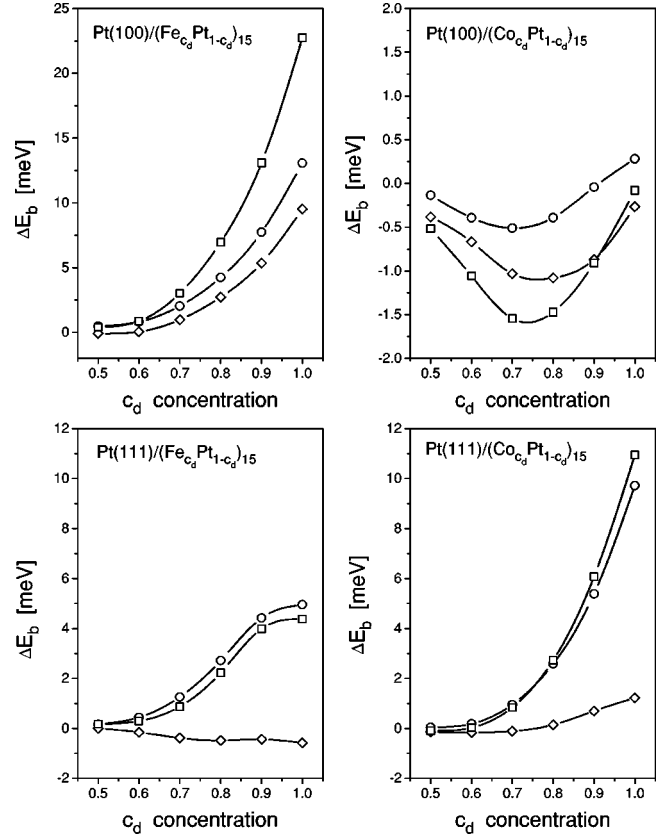In Fig. 1 the magnetic anisotropy energy of Pt terminated

FIG. 2. Band energy contribution (squares) to the magnetic anisotropy energy with respect to increasing ordering (interdiffusion parameter c_d) for a 15-layer-thick Fe/Pt (left) and Co/Pt (right) film on Pt(100) (top) and Pt(111) (bottom). Also shown are the corresponding Fe (circles) or Co (circles) and Pt (diamonds) contributions.

(FePt) superstructures on top of Pt(100) and Pt(111), see also Table I, is shown together with ΔE_b and ΔE_{dd} as a function of the number of repetitions. As can be easily seen, in both cases a perpendicular orientation of the magnetization is preferred. The magnetic anisotropy energy per repetition is much bigger in the case of a Pt(100) substrate than for a Pt(111) substrate. It should be noted that only ΔE_a per repetition (not shown in Fig. 1) tends to a constant for a large enough number of repetitions, see in particular also Refs. 15 and 16.

As to be expected the magnetic anisotropy energy is very sensitive to possible interdiffusion at the interfaces of the $3d$ metal with Pt. Assuming an interdiffusion model as defined in Table I, interesting differences between the two kinds of substrates on the one hand, and Co and Fe on the other hand can be seen (Fig. 2). In the case of interdiffused (FePt) superstructures on Pt(111) the Pt-like contribution to ΔE_b is negative, while for a Pt(100) substrate this contribution is positive and increasing notably with increasing ordering in the superstructure. In comparing the Fe-related results in Fig. 2 with those in Fig. 1, one has to keep in mind that — as already mentioned — the total number of atomic layers n corresponds to $r/2$, where r is the number of repetitions. The data in Fig. 2 refer, therefore, in Fig. 1 to the values for $r = 7$. As easily can be seen, in the present case the type of terminations has only little influence on the actual size of the magnetic anisotropy energy.
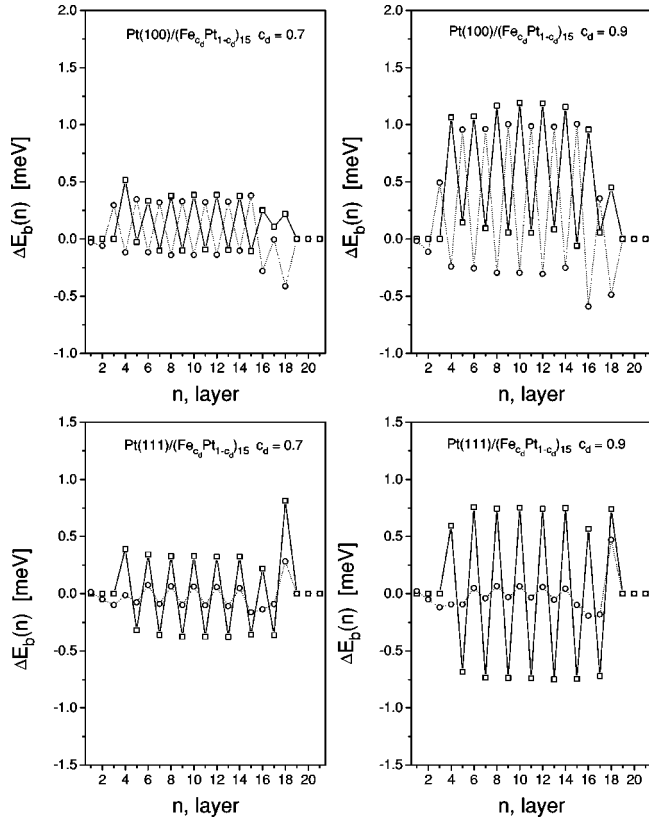
FIG. 3. Layer- and component-resolved band energy contributions to the magnetic anisotropy energy for interdiffused Fe/Pt films, 15 ML thick, on Pt(100) (top) and Pt(111) (bottom). The interdiffusion parameter c_d is 0.7 (left) and 0.9 (right). Note that the Fe-like (squares) and Pt-like (circles) contributions are not weighted by their respective layer concentrations.
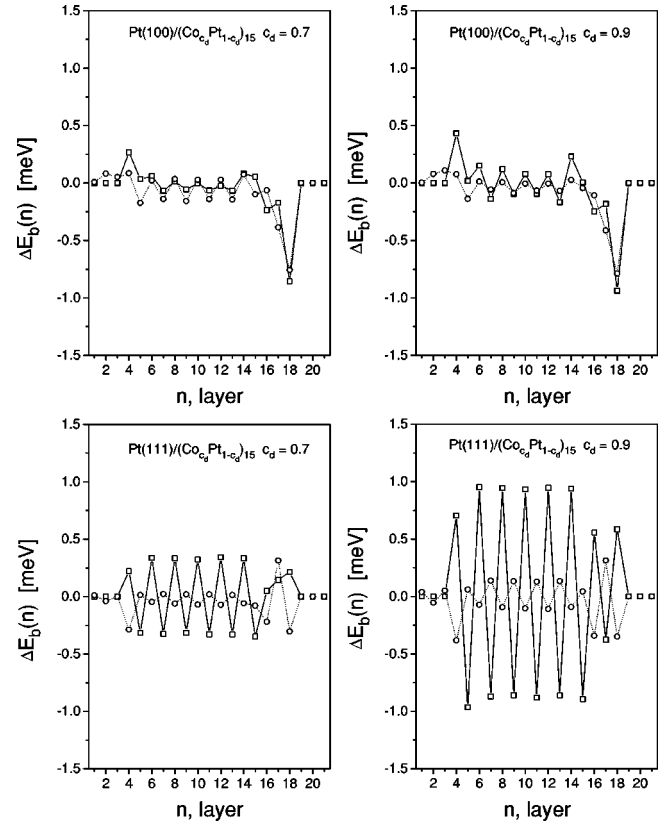
FIG. 4. Layer- and component-resolved band energy contributions to the magnetic anisotropy energy for interdiffused Co/Pt films, 15 ML thick, on Pt(100) (top) and Pt(111) (bottom). The interdiffusion parameter c_d is 0.7 (left) and 0.9 (right). Note that the Co-like (squares) and Pt-like (circles) contributions are not weighted by their respective layer concentrations.

For interdiffused (CoPt) superstructures on a Pt(111) substrate such an increase in the Pt-like contribution is only experienced for $c_d \geqslant 0.8$. Up to this value ΔE_b is about the same for Fe- as well as Co-derived interdiffused superstructures on Pt(111), i.e., cannot be related simply to either the componentlike (spin-only) magnetic moments for Fe or Co. Interdiffusion of (CoPt) superstructures on Pt(100) immediately causes the magnetization to reorientate parallel to the surface. As can be seen from Fig. 2, both the Co- and the Pt-like contributions are negative, the Co-like contribution changing sign only close to the ordered structure with ΔE_b remaining negative. This is not surprising since even for the corresponding Pt terminated superstructure with eight repetitions,[9] ΔE_b is only about 1.8 meV. The comparison between the two surface orientations in Fig. 2 quite impressively demonstrates that the differences to be seen are not caused simply by the different interlayer spacing, namely, 3.706 85 a.u. and 4.280 30 a.u. for the (100) and the (111) surface direction of a parent fcc Pt lattice, respectively, but by the very details of the electronic structure.

The substantial difference between the two surface orientations becomes immediately clear when analyzing component- and layer-resolved band energy differences, see Eq. (3). Although for both types of surfaces oscillations with a period of two, the thickness (in ML) of the repeated unit, seems to be characteristic, it is essentially the Pt-like contribution that is distinctly different: for the Fe-related systems

in Fig. 3 with (111) surface orientation the Pt-like layer-resolved band energy contributions are "in phase" with the Fe-like contributions, while for the (100) orientation they are "out of phase." Furthermore, in the case of the (111) orientation the Fe-like contributions from Pt-rich layers are negative, whereas for (100) all Fe-like contributions are positive. With increasing degree of ordering the $\Delta E_{b,p}^{\alpha}$ grow in size, keeping, however, their characteristic patterns. For the Co-related systems (Fig. 4) a different situation occurs: as compared to the corresponding Fe-related system for the (111) orientation the amplitudes of the Pt-like contributions oscillate "out of phase" with those of Co. The most dramatic effects are caused by the surface and the subsurface atomic layer. For a Pt(100) substrate the Co as well as the Pt contribution from these two layers is by far biggest and negative, see also Fig. 2. Even for the Pt(111) substrate the perturbations near the surface are quite notable. It should be noted that in accordance with Eq. (3), in Figs. 3 and 4 the component- and layer-resolved band energy differences are not weighted by their respective layer concentrations. As all systems discussed refer to free surfaces the last three entries correspond to vacuum layers: the respective Fe- (Co-) and Pt-like layer-resolved band energy contributions in these layers are exactly zero. In all cases the first three layers are pure Pt layers (see also Table I), which serve as "buffers" to the semi-infinite substrate.

In the present study we have shown quantitatively that the

magnetic anisotropy energy of (Fe/Pt) and (Co/Pt) super-structures on Pt(100) and Pt(111) is extremely sensitive to interdiffusion and thus confirmed the qualitative experience found in various experimental investigations. Furthermore, we have pointed out the importance of the Pt-like contributions to ΔE_a and illustrated the differences between Fe and Co and between different orientations of the substrate. Since experimental samples very often are not in thermodynamical equilibrium, a direct comparison to experimental data is only useful in cases where well-characterized systems are reported. Quite clearly aside from different concentration profiles to be considered and different types of superstructures with different repeat units [M_mPt$_n$ (M = Fe,Co)], in addition to interdiffusion phenomena relaxation effects most likely occur. In principle they can be included[12] into *ab initio* studies like the present one, but need to be guided by experi-mental data. The necessity, however, to use experimental techniques to stabilize ordered superstructures in order to achieve a maximum perpendicular magnetic anisotropy, i.e., an optimization of magneto-optical properties,[17] is quite impressively already demonstrated with the present theoretical study.

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