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Origin of orbital magnetization and magnetocrystalline anisotropy in *TX* ordered alloys (where T=Fe,Co and X=Pd,Pt)

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We present a method to calculate the magnetic anisotropy parameters which is based on a perturbative treatment of the spin-orbit interaction and a Green's function technique in real space. It allows us to interpret the magnetocrystalline anisotropy energy (MAE) in terms of interatomic interactions in the crystal. The method is applied to analyze orbital magnetism and MAE in *TX* ordered alloys (T=Fe,Co and X=Pd,Pt). The convergence of the orbital moments and MAE in real space and its relation to the problems of Brillouin-zone integration and of oscillatory behavior of MAE as a function of band filling are discussed. A comparison with results obtained by other methods is also given.

I. INTRODUCTION

During the last years a lot of experimental and theoretical work has been devoted to investigating orbital magnetism and magnetocrystalline anisotropy of ferromagnetic materials. This has been spurred on by the synthesis of a large number of new artificial systems with lower symmetry (such as overlayers, multilayers, interfaces) and the search for new promising candidates for magnetic recording materials and other applications. Considerable progress in the theoretical description of these phenomena is related to the implementation of band structure methods which give the possibility to connect the considered properties with peculiarities of the electronic spectra and thus to understand the underlying physics on a microscopic level. Both semiempirical tight-binding[1,2] as well as *ab initio* electronic structure methods[3–15] have been used to investigate orbital magnetism and magnetocrystalline anisotorpy energy (MAE) in a variety of layered compounds. At present, band structure schemes based on the local-spin-density approximations (LSDA) and including the spin-orbit interaction either as a perturbation[3–10,13] or in the scope of a fully relativistic formalism[14,15] have already become more or less standard tools for such calculations. Very extended studies of the MAE phenomena have been done based on the **k**-space analysis and using conventional band structure methods. At the same time, for many fundamental questions concerning magnetic anisotropy and orbital magnetism in itinerant electron compounds, a physical picture based on interatomic interactions in real space seems to be more appropriate. Some of these are the following: How far are these effects extended in space? Are they pure "single-site" properties and can they be considered as a result of interplay between spin polarization, spin-orbit interaction, and crystal field splitting on locally the same site? Or does the direct interatomic hybridization in combination with the spin-orbit interaction play an essential role in forming the orbital magnetization and the magnetocrystalline anisotropy? In this respect we would like to recall that for a long time the orbital magnetism was attributed entirely to systems with localized electrons. Thus, it is strongly desired to formulate a scheme which allows us to express the orbital magnetization and MAE in terms of contributions from different sites (groups of sites) and interatomic interactions in real space. Some preliminary progress in this field has been achieved in Ref. 9, where first-principles MAE results for two-dimensional systems were explained in terms of very simplified diatomic-pair models. The present paper is devoted to a solution of this problem. We start with a scalar-relativistic Green's function technique in real space and show that in lowest order perturbation theory with respect to the spin-orbit interaction (SOI) the orbital magnetization and MAE can be expressed in the required form. The idea of such a treatment of the interatomic interactions is well known in the multiple-scattering formalism and was intensively exploited for *ab initio* exchange calculations in a variety of compounds.[16] A very similar formalism based on the fully relativistic spin-polarized approach and considering the deviation of magnetization from equilibrium on every site as a perturbation has been developed by Antropov and Liechtenstein[17] and independently by Staunton *et al.*,[18] where it was also used to examine the anisotropic Ruderman-Kittel-Kasuya-Yosida interaction between two impurities in an electron gas.

The method presented in Sec. II will be applied to study the magnetic anisotropy of the *TX* ordered alloys (where T=Fe,Co and X=Pd,Pt). These layered compounds (crystallizing in the AuCu structure with alternating monolayers of T and X atoms) exhibit extremely large values of the perpendicular MAE (Ref. 19) (comparable to those for some rear-earth compounds). Particular attention will be paid to the convergence of the considered properties in real space and a comparison with results obtained by other methods. In Sec.

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IV we will give a short conclusion and illustrate how the convergence problem of the orbital moments and MAE in real space is connected with the number of **k** points required for the Brillouin-zone integration and with the oscillatory behavior as a function of band filling. Two appendixes are devoted to the single-impurity approximation for the SOI and the orbital polarization effect in the context of magnetic anisotropy calculations.

II. METHOD

The orbital moment induced on the ith site by spin-orbit interaction can be expressed through the on-site Green's function $G^{ii}(E)$ as

$$M_L^i = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F} dE\ \mathrm{Tr}\{L_\| G^{ii}(E)\}, \tag{1}$$

where $L_\|$ is a projection of the orbital moment on the spin magnetization direction, and Tr runs over the spin and orbital variables. If we start with the scalar-relativistic formalism and consider the SOI as a perturbation, the full Green's function $G(E)$ is related to the scalar-relativistic one $\tilde{G}_0(E)$ by Dyson's equation:

$$G(E) = \tilde{G}_0(E) + \tilde{G}_0(E)\Delta V(E) G(E). \tag{2}$$

The SOI $\Delta V(E)$ which is diagonal in the site indices and, for spherical potentials, also in the orbital quantum numbers can be expressed in the well-known form

$$\Delta V^i_\ell(E) = \xi^i_\ell(E)\mathbf{LS}, \tag{3}$$

where the spin-orbit coupling parameters $\xi^i_\ell(E)$ can be expressed by the radial derivatives of the potential V_i inside the ith Wigner-Seitz sphere (S_i) and the radial wave functions as

$$\xi^i_\ell(E) = \frac{1}{c^2}\int_0^{S_i} r R^2_\ell(r,E)\frac{dV_i(r)}{dr}dr \tag{4}$$

(c is the light velocity). The scalar-relativistic Green's function $\tilde{G}_0(E)$ for an arbitrary orientation of spin magnetization $\mathbf{e} = (\sin\vartheta\cos\varphi, \sin\vartheta\sin\varphi, \cos\vartheta)$ can be obtained from the one for the magnetization in z direction $\mathbf{e} = (0,0,1)$ by a unitary transformation $U_S(\vartheta,\varphi)$ in spin subspace

$$\tilde{G}_0(\vartheta,\varphi) = U_S^{-1}(\vartheta,\varphi) G_0 U_S(\vartheta,\varphi), \tag{5}$$

where

$$U_S(\vartheta,\varphi) = \begin{pmatrix} \cos\vartheta/2 & \sin\vartheta/2 e^{-i\varphi} \\ -\sin\vartheta/2 e^{i\varphi} & \cos\vartheta/2 \end{pmatrix}$$

are Wigner's rotation matrices. Then, within the first order of perturbation theory M_L^i can be written as sum over contributions from sites j,

$$M_L^i = \sum_j M_L^{ij}, \tag{6}$$

where M_L^{ij} is given by

$$M_L^{ij} = -\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F} dE\ \mathrm{Tr}\{L_\| \tilde{G}_0^{ij}\Delta V^j \tilde{G}_0^{ji}\}. \tag{7}$$

Here we use that in absence of SOI the orbital moment is quenched. M_L^{ij} can be interpreted as the contribution to the orbital moment on site i being induced by the spin-orbit coupling ΔV^j on site j. In order to evaluate the MAE we start with the single-particle energies

$$E_0 = \int_{-\infty}^{E_F} dE\ E n(E) - E_F\{N(E_F) - N\}. \tag{8}$$

Here $n(E)$ is the density of states and $N(E)$ the corresponding integrated density of state. The second part in Eq. (8) has the meaning of a Lagrangian constraint which has been added to ensure the extremal properties of the total energy for non-charge-conserving variations. N is the exact number of electrons. By partial integration E_0 is obtained as

$$E_0 = E_F N - \int_{-\infty}^{E_F} dE\ N(E). \tag{9}$$

Thus the change of the single-particle energies due to SOI is given by

$$\varepsilon = -\int_{-\infty}^{E_F} dE \delta N(E). \tag{10}$$

According to Lloyd's formula the change of the integrated density of states is evaluated as

$$\delta N(E) = -\frac{1}{\pi}\mathrm{Im}\ \mathrm{Tr}\ \ln(1 - \Delta V \tilde{G}_0). \tag{11}$$

Then, in second-order perturbation theory with respect to the SOI (due to time-reversal symmetry for the unperturbed system the first-order contribution vanishes), ε can be expressed as a sum of two-site interactions

$$\varepsilon = \sum_{ij} \varepsilon^{ij}, \tag{12}$$

with

$$\varepsilon^{ij} = -\frac{1}{2\pi}\mathrm{Im}\int_{-\infty}^{E_F} dE\ \mathrm{Tr}\{\tilde{G}_0^{ij}\Delta V^j \tilde{G}_0^{ji}\Delta V^i\}. \tag{13}$$

The angular dependence of the orbital moment and the MAE obeys the symmetry of the considered system. For O_h symmetry M_L^i and ε do not depend on the orientation of magnetization at all (in second-order perturbation theory). For crystals with uniaxial symmetry M_L^i and ε exhibit the sine-square dependence on the orientation of magnetization with respect to the perpendicular axis (ϑ),

$$M_L^i(\vartheta) = M_L^i(0) + \Delta M_L^i \sin^2\vartheta, \tag{14}$$

and correspondingly,

$$\varepsilon(\vartheta) = \varepsilon(0) + \Delta\varepsilon \sin^2\vartheta. \tag{15}$$

Analytical expressions for $M_L^i(\vartheta)$ and $\varepsilon(\vartheta)$ obtained in first and second orders of the regular perturbation theory can

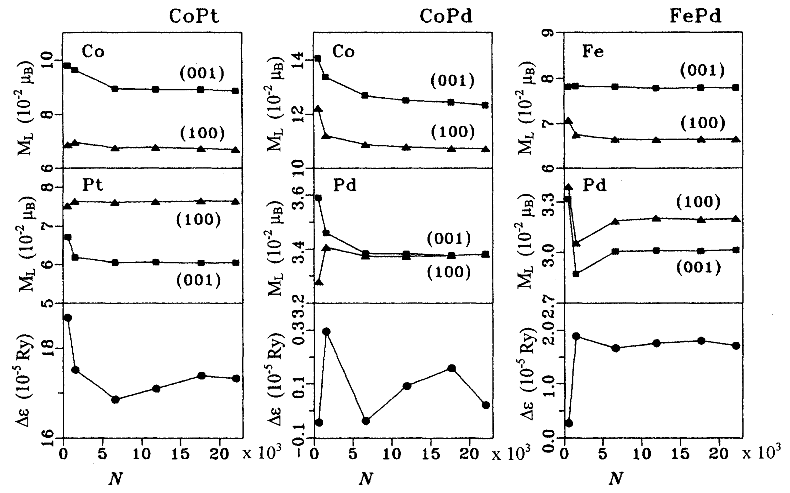

FIG. 1. Calculated orbital moments for two directions and MAE as a function of the number of **k** points in the full Brillouin zone for CoPt, CoPd, and FePd ordered alloys.

be found in Ref. 1. The tetragonal symmetry case is discussed in Appendix A within the scope of the so-called "single-impurity" approximation where the orbital magnetization is considered solely as a response to the SOI on the same site. The dependence on the in-plane rotations (φ) can appear only in higher orders of perturbation theory. Using expressions (6) and (7) and (12) and (13) the perpendicular anisotropy parameters $\Delta M_L^i = M_L^i(\pi/2) - M_L^i(0)$ and $\Delta\varepsilon = \varepsilon(\pi/2) - \varepsilon(0)$ can be also decomposed in partial contributions arising from different sites of the crystal. But it is necessary to note here that the angular dependence for the separate components of the orbital moment M_L^{ij} and magnetocrystalline anisotropy energy ε^{ij} is not the same since only the total values (14,15) or combinations such as

$$M_L^{0\alpha} = \sum_{i \in \{g_\alpha\}} M_L^{0i}, \tag{16}$$

where the summation extends over all equivalent sites, obey Eq. (14). (The analogous expression is valid for ε^{ij}.)

Finally, using the arguments described above, the MAE of a system consisting of two (or more) types of atoms (say T and X) can be expressed as a sum

$$\Delta\varepsilon = \Delta\varepsilon^T + \Delta\varepsilon^X, \tag{17}$$

where

$$\Delta\varepsilon^{T(X)} = \sum_i \Delta\varepsilon^{T(X)i} \tag{18}$$

is the partial contribution related to a given type of atom. For layered compounds one can also introduce layer-dependent contributions to the orbital magnetization and MAE due to layers located at different distances from the considered atom,

$$\Delta\varepsilon^{\|} = \sum_{i \in \text{layer}} \Delta\varepsilon^{0i}. \tag{19}$$

(The analogous equation holds for the orbital magnetization.)

III. RESULTS AND DISCUSSIONS

A. Details of calculations and simplifications

In the following we present our results for the layered TX alloys with AuCu structure. Using Eqs. (6) and (12) the orbital magnetization and magnetic anisotropy parameters can be calculated by direct summation in real space. Practically, the spin-orbit interaction has been switched on only for states of d symmetry in a cluster consisting of 791 atoms on 76 nonequivalent sites. The interatomic distances from the center of the cluster (actually we considered two different clusters surrounding T and X atoms) do not exceed $5a$, where a is the distance between nearest neighbors in the planes of equivalent atoms. The scalar-relativistic Green's function G_0 has been obtained using the linear muffin-tin orbital[20,21] (LMTO) method in the nearly orthogonal representation.[22,23] All calculations have been performed with the experimental values of the lattice parameters, as reported in Refs. 5 and 24. The parameters of CoPt have also been used for the hypothetical compound CoPd. The Wigner-Seitz sphere radii have been chosen from the charge neutrality condition for every component of the alloys. For details and results of regular band structure calculations for the considered compounds we refer to Ref. 15 (see also Refs. 5, 24, and 25).

The most important question in calculations of the magnetocrystalline anisotropy is the convergence of the Brillouin-zone (BZ) integrals. We display in Fig. 1 the orbital moments for two different orientations and the MAE as a function of **k**-space filling obtained for CoPt, CoPd, and FePd ordered alloys in LMTO formalism where the SOI was included in a "pseudoperturbation" manner.[20,28] Already 6615 **k** points corresponding to 20 divisions along the in-plane and 12 divisions along the perpendicular reciprocal-lattice vectors in a Γq lattice[29] give reasonably good convergence in the case of CoPt, FePd, and FePt (not shown in Fig. 1) alloys if the tetrahedron method[26] with correct weights for the tetrahedra is used.[27] Much more accuracy is needed to reproduce the relatively small values of M_L^{Pd} and MAE parameters for a hypothetical CoPd alloy and even a mesh with 22 103 **k** points (corresponding to 30 and 22 divisions of reciprocal-lattice vectors) appears to be insufficient. Results illustrating the real-space expansion of orbital magnetization,

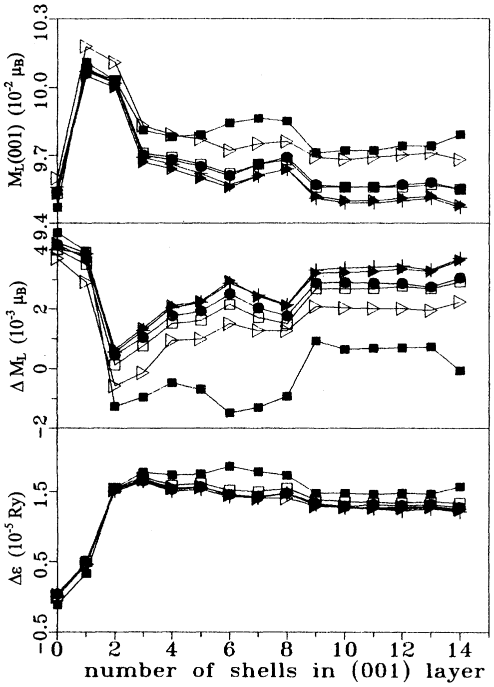

FIG. 2. Summation in the real space performed for an (001) layer of Co atoms in CoPt with different number of **k** points in Brillouin zone: 567 (black squares), 1521 (white triangles), 6615 (white squares), 11 875 (black circles), 17 661 (black triangles), and 22 103 (crosses).

its anisotropy, and MAE obtained for a Co(001) layer in CoPt [Eq. (19)] are shown in Fig. 2. The different curves correspond to different numbers of **k** points in the BZ which have been used in the calculation of the Green's function $\tilde{G}_0$. One can see again that a choice of 6615 **k** points in the whole BZ is quite reasonable for this system and reproduces all main details of the picture.

From the physical point of view the number of **k** points required in MAE calculations strongly depends on the ratio between "surface" and "volume" components of MAE. Kondorskii and Straube have shown[30] that quasidegenerate states near the Fermi surface give the main contribution to the MAE for ferromagnetic Ni (the contributions to the MAE from other regions in the Brillouin zone were shown to be several orders of magnitude smaller). This conclusion was supported in the following by first-principles calculations of the MAE for Fe, Co, and Ni performed by Daalderop *et al.*,[31] and Guo *et al.*,[32] where only extremely fine mesh in reciprocal space (about 10^6 **k** points in the Brillouin zone) gave a reasonable convergence for the "surface" contribution to the MAE. The situation is probably different for systems where due to the lowering of the crystalline symmetry the magnetic anisotropy effects appear in lower order with respect to the SOI. In second order of perturbation theory (we would like to recall again here that for cubic metals the MAE effect first appears in fourth-order perturbation theory), the change of the one-electron energy induced by the SOI is given by

$$\varepsilon=-\sum_{\mathbf{k}}\sum_{n}^{\mathrm{occ}}\sum_{m}^{\mathrm{unocc}}\frac{|\langle m\mathbf{k}|\xi\mathbf{LS}|n\mathbf{k}\rangle|^2}{E_m(\mathbf{k})-E_n(\mathbf{k})}. \tag{20}$$

For every **k** point it decays only as the inverse energy difference between occupied and empty states and contributions from the single-particle states rather far from the Fermi surface can also be appreciable. Thus, one can expect that for uniaxial systems the "volume" part of the MAE is significantly enhanced in comparison to the "surface" one. Actually this fact was intensively used in the so-called state-tracking approach proposed by Wang *et al.*,[8] which shows rather fast convergence in **k** space but is based on a very approximate treatment of the states close to the Fermi surface. This method gives quite reasonable results for a series of magnetic multilayers[9–11] but converges to the wrong MAE for fcc-Ni (Ref. 33), known as a typical system where the "surface" contribution is very important. Generally, the regular perturbation theory expression (20) is valid if $|E_m(\mathbf{k})-E_n(\mathbf{k})|\gg|\langle m\mathbf{k}|\xi\mathbf{LS}|n\mathbf{k}\rangle|$ (otherwise the quasidegenerate perturbation theory should be used). Therefore, it is not applicable for the quasidegenerate pairs close to the Fermi surface. This is a general problem for the perturbative treatment of the SOI no matter whether the regular expression (20) or the Green's function technique described in the previous section is used.[34] In order to justify the validity of this approach for the *TX* ordered alloys we have performed calculations where the SOI was included in two different ways. The first one is the regular perturbation theory where the change of one particle energies is given by Eq. (20). A corresponding expression for the induced orbital magnetization can be easily found using the first-order perturbation theory for the wave functions. All contributions from quasidegenerate pairs close to the Fermi surface have been neglected. Practically, we enforced matrix elements of the SOI between states m and n to be zero if the condition $|E_m(\mathbf{k})-E_n(\mathbf{k})|<\delta_{\mathrm{cut}}$ has been fulfilled. The energy cutoff δ_{cut} is especially important in this approach. With small δ_{cut} ($\sim10^{-6}$ Ry), we found that the error from inappropriate treatment of the surface pair coupling is very large and the MAE was typically overestimated by several orders of magnitude (even if the quasidegeneracy occurs in a very restricted region of the BZ, this effect cannot be treated properly with a finite mesh of **k** points which introduces an unpredictable uncertainty in the numerical scheme). For $\delta_{\mathrm{cut}}=|\langle m\mathbf{k}|\xi\mathbf{LS}|n\mathbf{k}\rangle|$ the second-order correction for the one-electron energy (20) shows a very stable behavior. As a test we also performed calculations using a constant cutoff $\delta_{\mathrm{cut}}=5$ mRy and found results to be very similar to the previous choice. Thus, the effects attributed to the Fermi surface (both its deformation and coupling of the quasidegenerate states due to the SOI) are completely ignored in this approach. The second method is the so-called pseudoperturbation treatment,[20,28] where the matrix of SOI calculated on the scalar-relativistic basis functions was added to the band Hamiltonian explicitly. The eigenvalues were found by direct diagonalization of the Hamiltonian. For nearly crossing bands this procedure is equivalent to the quasidegenerate perturbation theory. Moreover, the position of the Fermi energy was redefined for every orientation of magnetization. Thus, this scheme is the most rigorous one and allows us to

TABLE I. Orbital moments $M_L(0)$ (in μ_b), anisotropy of orbital moments ΔM_L (in $10^{-3}\mu_b$), and MAE $\Delta\varepsilon$ (in 10^{-5} Ry) calculated for the TX ordered alloys on the basis of different schemes: pseudoperturbation treatment where the spin-orbit interaction was included for all states (PPT$_1$), for $T(d)$, $X(d)$ and $X(p)$ states (PPT$_2$), only for $T(d)$ and $X(d)$ states (PPT$_3$), and for $T(d)$ and $X(d)$ states through energy-independent spin-orbit coupling parameters (PPT$_4$); regular perturbation theory (PT) where the structure of the SOI is analogous to PPT$_4$.

Compound	Method	$M_L^T(0)$	ΔM_L^T	$M_L^X(0)$	ΔM_L^X	$\Delta\varepsilon$
FePt	PPT$_1$	0.0739	1.3	0.0444	11.6	24.8
	PPT$_2$	0.0735	2.1	0.0446	11.8	25.5
	PPT$_3$	0.0792	12.7	0.0451	8.6	21.1
	PPT$_4$	0.0789	-7.6	0.0426	7.9	10.5
	PT	0.0896	-4.6	0.0476	12.0	13.4
FePd	PPT$_1$	0.0781	-11.7	0.0301	1.8	1.7
	PPT$_2$	0.0784	-11.5	0.0298	2.4	2.1
	PPT$_3$	0.0821	-10.9	0.0296	0.9	0.4
	PPT$_4$	0.0782	-8.8	0.0271	0.7	0.4
	PT	0.0841	-9.8	0.0292	1.1	0.4
CoPt	PPT$_1$	0.0895	-22.0	0.0605	15.6	16.8
	PPT$_2$	0.0885	-21.5	0.0606	15.9	17.1
	PPT$_3$	0.0796	-32.8	0.0624	13.5	17.2
	PPT$_4$	0.0829	-22.5	0.0577	11.4	8.6
	PT	0.0996	-30.8	0.0632	15.8	12.0
CoPd	PPT$_1$	0.1270	-18.2	0.0338	-0.1	<0
	PPT$_2$	0.1261	-18.0	0.0337	>0	0.1
	PPT$_3$	0.1255	-21.5	0.0339	-0.7	0.3
	PPT$_4$	0.1186	-17.0	0.0307	-1.3	0.7
	PT	0.1369	-15.7	0.0270	7.8	1.2

consider both "volume" and "surface" effects in the MAE calculations on an equal footing. Results of such calculations are listed in Table I. One can see that for most compounds the regular perturbation theory gives already reasonable values for the orbital moments, their anisotropy, and MAE (lines PT and PPT$_4$ in Table I should be compared since they correspond to the same form of the SOI operator). Probably the situation is critical only for the hypothetical CoPd alloy where the regular perturbation theory predicts the wrong sign for the anisotropy of the orbital magnetization at Pd sites and overestimates the magnitude of MAE by a factor of 2. A similar effect has been found for a Pd/Co/Pd sandwich where the surface pair coupling was reported[10] to be negative and of the same order as the volume contribution to the MAE.

The role of the SOI for states with angular momenta other than d, which were omitted in Green's function analysis, and their influence on the distribution of orbital moments and magnitude of MAE can be shown by the following example. The main contribution to the integrals (4) arises from the region very close to the origin where the radial wave functions R_ℓ are essentially determined by the centrifugal term $\ell(\ell-1)/r^2$. Then, due to the behavior $R_\ell \sim r^\ell$ the states with smaller ℓ should have a larger weight in this region and the SOI parameters ξ_ℓ are expected to decrease with increasing ℓ. Indeed, for 5d and 6p states of Pt atoms we have estimated ξ_ℓ in the centers of occupied bands correspondingly as 42 and 163 mRy. Thus, in combination with large hybridization between d and p states of neighboring sites, the effect of SOI on Pt(6p) states can be appreciable. For FePt this interaction is especially important for the formation of the orbital moment on Fe sites and its anisotropy (Table I).

Finally, in all calculations based on the Green's function technique we have neglected the energy dependence of the spin-orbit coupling parameters $\xi(E)$ and replaced them by the values estimated in the centers of the occupied bands. We expect this approximation to be quite reasonable and to reproduce all features of the considered parameters (Table I).

B. Real-space analysis of orbital magnetization and MAE

Let us first consider the single-site components of the orbital moments M_L^{00}, which can be found using the results of Appendix A. The general tendencies of appearance of M_L^{00} on T and X sites can be well understood in terms of the exchange splitting and the magnitude of the SOI. In first-order perturbation theory only spin-diagonal elements of the SOI $\xi L_z S_z$ contribute to the orbital magnetization. Obviously, both spin polarization and SOI are important to unquench the orbital moment. On the one hand, the effect is proportional to the strength of SOI ξ. On the other hand, the orbital contributions induced by the SOI in different spins channels have opposite signs. If the occupations were equivalent for both spins a perfect cancellation would take place. Since due to exchange splitting the states for one spin direction will be preferentially occupied, the cancellation between different spins will be partially broken and an orbital magnetization obeying Hund's third rule is formed. Thus, the size of the orbital moment depends on the spin splitting. Larger spin polarization is important for unquenching of orbital moments at 3d sites. Indeed, all of them are formed mainly by the states with spin down and renormalization by nearly occupied spin-up band is very poor (Table II). For Pd

TABLE II. Spin moments M_S, orbital moments $M_L(0)$ (in μ_b), and anisotropy of orbital moments ΔM_L (in $10^{-3}\mu_b$) obtained in the single-impurity approximation for spin-orbit interaction (contributions of different spin directions are shown in parentheses).

Compound	M_S^T	$M_L^T(0)$	ΔM_L^T	M_S^X	$M_L^X(0)$	ΔM_L^X
FePt	2.77	0.0580	-11.7	0.35	0.0553	10.0
		(-0.0076,0.0656)	(-0.5,-11.2)		(-0.1126,0.1679)	(30.6,-20.6)
FePd	2.86	0.0649	-9.1	0.35	0.0412	-6.7
		(-0.0058,0.0707)	(0.2,-9.3)		(-0.0229,0.0641)	(0.8,-7.5)
CoPt	1.72	0.0953	4.0	0.37	0.0730	17.1
		(-0.0107,0.1060)	(-1.0,5.0)		(-0.1110,0.1840)	(30.0,-12.9)
CoPd	1.81	0.1113	15.1	0.36	0.0485	-4.6
		(-0.0085,0.1198)	(0.1,15.0)		(-0.0244,0.0729)	(2.2,-6.8)

and Pt sites the exchange splitting is much weaker and a strong cancellation between orbital magnetizations induced in different spin channels takes place. On the other hand, the SOI parameters are several times larger for heavy Pd and Pt atoms (the ξ parameters estimated in the center of gravity of the occupied d band are 4, 5, 15, and 42 mRy for Fe, Co, Pd, and Pt, respectively), which considerably enhances the orbital moments and makes them comparable to those at T sites. Also larger M_L^{00} on Pt sites is caused by the larger SOI. The behavior of the orbital magnetization at T sites due to substitution of Fe by Co sublattices is more complicated and mainly related with distribution of spin-down $3d$ states of xy and x^2-y^2 symmetry near the Fermi level. Coupling between these states [first term in Eq. (A1)] is most important and responsible for ~85% of orbital magnetization unquenched at T sites.

The single-site terms M_L^{00} give the main contributions to the orbital moments for all sites of the considered systems. In contrast to this the anisotropy of the orbital magnetization appears to be less localized in space and its ΔM_L^{00} component is of the same order as contributions from the nearest spheres of neighbors. The real-space convergence of the orbital magnetization and MAE within one Co(001) layer in the CoPt alloy was already considered in the previous section (Fig. 2) in the context of BZ integration. One can see that about 15 shells of atoms in the square lattice are sufficient to reproduce the total single-layer contribution to the orbital magnetization and MAE. We do not show here analogous results obtained for other systems, which will be published elsewhere. In general, our experience shows that for all layers in the considered compounds such a number of shells in the planes guarantees a reasonable in-plane convergence. However, in many cases the convergence can be faster than this. For example, already the single-site component and two spheres of nearest neighbors in Pd layers give a reasonable convergence for the contribution of a single layer to the or-

TABLE III. Contributions of different layers to the orbital moments $M_L^{\parallel}(0)$ (in μ_b), anisotropy of orbital moments $\Delta M_L^{\parallel}$ (in $10^{-3}\mu_b$), and MAE $\Delta\varepsilon^{\parallel}$ (in 10^{-5} Ry) for Fe and Pt sites in FePt ordered alloy.

Site	Layers	$M_L^{\parallel}(0)$	$\Delta M_L^{\parallel}$	$\Delta\varepsilon^{\parallel}$
	Fe(0,0,0)	0.0734	-18.1	2.6
	Pt(0,0,± c/2)	0.0069	3.1	-2.3
	Fe(0,0,± c)	-0.0007	0.5	-0.2
Fe	Pt(0,0,± 3c/2)	-0.0001	3.7	-1.1
	Fe(0,0,± 2c)	0.0003	-0.6	0
	Pt(0,0,± 5c/2)	0.0003	0.1	-0.1
	Fe(0,0,± 3c)	0	0	0
	Pt(0,0,± 7c/2)	0.0001	0.2	0
Fe	Total	0.0802	-11.2	-1.1
	Pt(0,0,0)	0.0529	11.0	14.9
	Fe(0,0,± c/2)	0.0006	0.3	-2.3
	Pt(0,0,± c)	-0.0051	1.4	8.7
Pt	Fe(0,0,± 3c/2)	0.0001	0.3	-1.1
	Pt(0,0,± 2c)	0	0.3	-2.1
	Fe(0,0,± 5c/2)	0	0	-0.1
	Pt(0,0,± 3c)	0.0001	-0.4	-0.2
	Fe(0,0,± 7c/2)	0	0	0
Pt	Total	0.0486	13.0	17.7

TABLE IV. Contributions of different layers to the orbital moments, anisotropy of orbital moments, and MAE for Fe and Pd sites in FePd ordered alloy (all notations as in Table III).

Site	Layers	$M_L^{\parallel}(0)$	$\Delta M_L^{\parallel}$	$\Delta\varepsilon^{\parallel}$
	Fe(0,0,0)	0.0788	-15.6	2.8
	Pd(0,0,± c/2)	0.0021	1.0	-0.7
	Fe(0,0,± c)	-0.0008	1.7	-0.1
Fe	Pd(0,0,± 3c/2)	-0.0002	1.0	-0.3
	Fe(0,0,± 2c)	0.0002	-0.9	0.1
	Pd(0,0,± 5c/2)	0	0	0
	Fe(0,0,± 3c)	0	0.1	0
	Pd(0,0,± 7c/2)	0	0.1	0
Fe	Total	0.0805	-12.5	1.7
	Pd(0,0,0)	0.0299	0.4	0.9
	Fe(0,0,± c/2)	0.0005	0.3	-0.7
	Pd(0,0,± c)	-0.0008	0.1	0.8
Pd	Fe(0,0,± 3c/2)	0.0001	0.3	-0.3
	Pd(0,0,± 2c)	0	-0.7	0.3
	Fe(0,0,± 5c/2)	0	0	0
	Pd(0,0,± 3c)	0	0	0
	Fe(0,0,± 7c/2)	0	0	0
Pd	Total	0.0296	0.4	0.9

TABLE V. Contributions of different layers to the orbital moments, anisotropy of orbital moments, and MAE for Co and Pt sites in CoPt ordered alloy (all notations as in Table III).

Site	Layers	$M_L^{\parallel}(0)$	$\Delta M_L^{\parallel}$	$\Delta\varepsilon^{\parallel}$
	Co(0,0,0)	0.0955	2.9	1.3
	Pt$(0,0,\pm c/2)$	0.0082	-9.2	-3.4
	Co$(0,0,\pm c)$	-0.0056	-4.7	0.7
Co	Pt$(0,0,\pm 3c/2)$	0	-6.1	-0.2
	Co$(0,0,\pm 2c)$	-0.0007	1.2	-0.2
	Pt$(0,0,\pm 5c/2)$	0	-0.7	-0.1
	Co$(0,0,\pm 3c)$	0	-0.1	0
	Pt$(0,0,\pm 7c/2)$	0	0	0
Co	Total	0.0976	-16.8	-1.9
	Pt(0,0,0)	0.0683	20.1	13.9
	Co$(0,0,\pm c/2)$	0.0010	-1.1	-3.4
	Pt$(0,0,\pm c)$	-0.0044	2.1	4.6
Pt	Co$(0,0,\pm 3c/2)$	0	-0.8	-0.2
	Pt$(0,0,\pm 2c)$	0.0001	0.5	1.1
	Co$(0,0,\pm 5c/2)$	0	-0.1	-0.1
	Pt$(0,0,\pm 3c)$	0.0001	-0.3	-0.9
	Co$(0,0,\pm 7c/2)$	0	0	0
Pt	Total	0.0651	20.3	15.0

TABLE VI. Contributions of different layers to the orbital moments, anisotropy of orbital moments, and MAE for Co and Pd sites in CoPd ordered alloy (all notations as in Table III).

Site	Layers	$M_L^{\parallel}(0)$	$\Delta M_L^{\parallel}$	$\Delta\varepsilon^{\parallel}$
	Co(0,0,0)	0.1246	0.8	1.3
	Pd$(0,0,\pm c/2)$	0.0016	-3.9	-0.6
	Co$(0,0,\pm c)$	-0.0083	-3.8	0.5
Co	Pd$(0,0,\pm 3c/2)$	0.0001	-2.8	0.1
	Co$(0,0,\pm 2c)$	0.0010	0.3	-0.1
	Pd$(0,0,\pm 5c/2)$	-0.0001	0	0
	Co$(0,0,\pm 3c)$	-0.0002	-0.5	0.1
	Pd$(0,0,\pm 7c/2)$	0	0	0
Co	Total	0.1187	-9.9	1.3
	Pd(0,0,0)	0.0362	2.1	0.3
	Co$(0,0,\pm c/2)$	0.0006	-1.4	-0.6
	Pd$(0,0,\pm c)$	-0.0025	1.6	-0.3
Pd	Co$(0,0,\pm 3c/2)$	-0.0001	-1.0	0.1
	Pd$(0,0,\pm 2c)$	0	0	0.1
	Co$(0,0,\pm 5c/2)$	0	0	0
	Pd$(0,0,\pm 3c)$	-0.0001	-0.1	0
	Co$(0,0,\pm 7c/2)$	0	0	0
Pd	Total	0.0341	1.2	-0.3

bital magnetization and its anisotropy in Pd-based ordered alloys, whereas an analogous expansion for the MAE converges much slower.

The contributions of different layers to the orbital moments and MAE for the *TX* ordered alloys are presented in Tables III–VI. Generally, the decay with distance in the (001) direction is not monotonous. Moreover, its behavior strongly depends on the type of layers due to the dependence on the SOI. For example, the *T*-*X* hybridization plays a very important role in the formation of the orbital moment anisotropy of lighter *T* elements, since in this case the hybridization effects are more important inducing a proportionality to the spin-orbit coupling parameters of the heavy Pd and Pt atoms. The negative values of ΔM_L^{Co} parameters in CoPt and CoPd alloys result mainly from the interaction with first and second neighboring Pt and Pd layers. In contrast to this, in Fe-based alloys the effects of *T*-*X* interaction on the anisotropy of the orbital magnetization on Fe sites are positive and partially compensate strongly negative contributions of the Fe layers passing through the origin. For orbital moments developed on *X* sites the interaction with 3*d* atoms appears only with the factor ξ^T and therefore corresponding contributions will be several times smaller.

The giant positive MAE for Pt-based compounds (the values of MAE parameters $\Delta\varepsilon^T+\Delta\varepsilon^X=\Delta\varepsilon$ calculated for CoPt and FePt ordered alloys are $-0.019+0.150=0.131$ and $-0.011+0.177=0.166$ mRy, respectively) can be entirely attributed to the Pt sublattice. As a first approximation for the MAE in these compounds one can assume that the lattice of magnetic 3*d* atoms acts as a "source of magnetism" only and induces the spin magnetization on Pt sites which due to the large SOI give the main contribution to the perpendicular magnetocrystalline anisotropy. The contributions of the 3*d* sites themselves $\Delta\varepsilon^T$ are relatively small and moreover have an opposite sign. The single-site terms $\Delta\varepsilon^{00}$ in the magnetocrystalline anisotropy parameters $\Delta\varepsilon^X$ for CoPt and FePt ordered alloys were found to be as large as 0.081 and 0.131 mRy. Interactions between Pt atoms lying in the same plane increase these values to 0.139 and 0.149 mRy, respectively. Then, the interaction with the two nearest Pt layers gives an additional positive contribution to the $\Delta\varepsilon^X$ (see Tables III and V). On the other hand, direct hybridization effects between atoms of different types located in neighboring layers were found to be negative (entirely due to this interaction the MAE on 3*d* sites $\Delta\varepsilon^T$ in Pt-based alloys is also negative). Thus, for the considered compounds the direct interaction between atoms of *T* and *X* types plays a destructive role in the perpendicular magnetocrystalline anisotropy formation.

As a result of the substitution of Pt atoms by the lighter Pd ones the absolute values of the MAE parameters of $\Delta\varepsilon^{XX}$ types are expected to decrease due to the difference in spin-orbit coupling constants for Pt and Pd by a factor $(\xi^{\mathrm{Pt}}/\xi^{\mathrm{Pd}})^2\approx 8$. Moreover, the *d* states of Pd atoms are known to be more localized in comparison with those of Pt. Therefore the decrease of the $\Delta\varepsilon^{XX}$ parameters should be even more pronounced (indeed, the contribution of one Pd layer to MAE in FePd constitutes only 0.009 mRy which is about 20 times smaller than the analogous value for a Pt layer in FePt). The absolute values for the mixed parameters $\Delta\varepsilon^{TX}$ also decrease from Pt- to Pd-based alloys, but this effect was found to be weaker since $\Delta\varepsilon^{TX}$ exhibit only a linear dependence on the ξ^X parameters. Thus, the balance between positive in-plane interactions of $\Delta\varepsilon^{TT}$ type and negative $\Delta\varepsilon^{TX}$ ones is shifted in Pd-based alloys in favor of $\Delta\varepsilon^{TT}$ and the MAE values $\Delta\varepsilon^T$ attributed to 3*d* atoms become positive. In FePd, both $\Delta\varepsilon^{\mathrm{Fe}}$ and $\Delta\varepsilon^{\mathrm{Pd}}$ are positive and enhance each other in the total MAE $\Delta\varepsilon=0.017+0.009=0.026$ mRy, whereas for the hypothetical CoPd alloy due to the sharp decrease of $\Delta\varepsilon^{XX}$ in comparison with $\Delta\varepsilon^{TX}$ the $\Delta\varepsilon^{\mathrm{Pd}}$ term changes its sign and leads to a partial cancellation between

$\Delta\varepsilon^{\mathrm{Co}}$ and $\Delta\varepsilon^{\mathrm{Pd}}$ contributions $\Delta\varepsilon=0.013-0.003=0.010$ mRy.

IV. SUMMARY AND CONCLUSIONS

We have presented a method to calculate the magnetic anisotropy based on a perturbation treatment for the SOI and a Green's function technique in real space. Despite its apparent laboriousness this approach gives an informative tool to analyze MAE calculations and allow us to interpret them in terms of interatomic interactions. The scaling factor for the angular and SOI strength dependence for uniaxial systems $\xi^2\sin^2\vartheta$ appears *analytically* in the MAE expression in our approach, thus, considerably facilitating the numerical problems (necessity to handle with small $\sim\xi^2$ energy differences and numerical fits) known in the standard MAE calculations. For the first time the method gives the possibility to investigate the convergence of the orbital magnetization and magnetic anisotropy in real space. Applications to the *TX* ordered alloys (where T=Fe,Co and X=Pd,Pt) have shown that a relatively small number of neighbors in real space carry the spin-orbit information, thus allowing us to calculate the orbital magnetization and the MAE being induced in the center of the cluster. This effect has two main consequences:

1. Due to oscillations of the exponential factor $\exp[i\mathbf{k}(\mathbf{R}_i-\mathbf{R}_j)]$ the number of $\mathbf{k}$ points required for accurate calculations of the Green's function elements G_0^{ij} increases with increasing distances $|\mathbf{R}_i-\mathbf{R}_j|$. Thus, due to the short-range behavior of orbital magnetization and MAE, the Green's function elements which give an appreciable contribution to the corresponding expressions [(6) and (12)] for these characteristics are essentially restricted by few spheres of nearest neighbors. This is in good agreement with the fact that even a moderately small number of $\mathbf{k}$ points allows us to reproduce reliable values for the orbital moments and anisotropy energies of the considered alloys.

2. Using tight-binding theory arguments, Heine and Samson[35] have shown that the oscillatory behavior of many physical variables, which can be expressed in terms of Green's functions, obeys as a function of band filling a general theorem. In particular, the larger the distance between the sites i and j, the more nodes the corresponding contributions to the MAE (13) should have (at least $2r$ in the tight-binding model, where r is a number of hops required to reach site j starting from i provided that only hops between nearest neighbors are allowed). Thus, since the main contributions to the MAE arise from interactions between sites located in the nearest neighborhood, one can expect that the number of nodes in the dependence on band filling is relatively small. The orbital magnetization, which is essentially determined by single-site components M_L^{00} (requiring no hop), should not change its sign at all when the Fermi level moves within a subband which is in agreement with Hund's third rule.

Direct hybridization between layers of T and X types (which together with the large spin-orbit coupling parameters of Pd atoms was expected[5,6] to give the main effect in perpendicular magnetic anisotropy of $\mathrm{Co_1Pd_2}$ multilayers) actually acts in favor of an in-plane orientation of the magnetization. It would be also interesting to investigate in the future the effects of hybridization between magnetic layers and spacers for different orientations and thicknesses of the multilayers. The source of the giant magnetocrystalline anisotropy in Pt-based alloys are interactions in the Pt sublattice (mainly in the layers and between nearest Pt layers), where the spin magnetization is induced by magnetic $3d$ sites.

Results of MAE calculations for the *TX* ordered alloys based on the Green's function technique are in reasonable agreement with those obtained by other (more traditional) methods taking into account the relativistic effects in band calculations, such as the pseudoperturbation treatment of the spin-orbit interaction (Table I, as well as the results of Ref. 5), or the spin-polarized fully relativistic approach.[15] In comparison with experimental data (MAE reported in Ref. 19 for FePt, CoPt, and FePd ordered alloys with equilibrium composition are 0.088, 0.061, and 0.021 mRy per formula unit, respectively), the values of MAE obtained in the present work are overestimated (by a factor of 2) for FePt and CoPt. For FePd we reach agreement with experiment considering the uncertainties of the method used in this work (Table I). One of the origins for the discrepancies in the Pt-based compounds could be the orbital polarization effects: exchange and correlations between electrons related with orbital degrees of freedom which are essentially beyond the uniform electron gas based LSDA description employed in the present work (a simple way to consider the orbital polarization effects based on a model Hartree-Fock approach is discussed in Appendix B). Also the temperature dependence of the magnetocrystalline anisotropy parameters could be very important (the present band structure calculations correspond to $T=0$, whereas all measurements have been performed at finite temperatures).

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APPENDIX A: "SINGLE-IMPURITY" APPROXIMATION FOR SPIN-ORBIT INTERACTION

Let us consider only single-site contributions to the orbital magnetization. From the formal point of view this approximation corresponds to the case where the spin-orbit interaction is neglected on all sites of the crystal except the considered one. That is why we denote it as "single-impurity." This approach has been used by Ebert *et al.*[36] and Shick *et al.*[37] to take relativistic effects in an impurity model based on the Green's function technique into account. For d states and tetragonal symmetry the corresponding scalar-relativistic Green's function G_0^{00} in the real harmonic basis is presented by four independent elements for every projection of spin σ: g_{xy}^{σ}, g_{xz}^{σ}, $g_{z^2}^{\sigma}$, and $g_{x^2-y^2}^{\sigma}$ (the element g_{yz}^{σ} is equal to g_{xz}^{σ} due to symmetry reasons). Then, for the orbital magnetization the following analytical expressions can be found:

$$M_L^{00}(0)=\xi\sum_{\sigma=\pm 1}\sigma\int^{E_F}\mathrm{Im}\{4g_{xy}^{\sigma}(E)g_{x^2-y^2}^{\sigma}(E)+(g_{xz}^{\sigma}(E))^2\}dE, \tag{A1}$$

$$M_L^{00}(\pi/2)=\xi\sum_{\sigma=\pm 1}\sigma\int_{-\infty}^{E_F}\mathrm{Im}\{g_{xz}^{\sigma}(E)[3g_{z^2}^{\sigma}(E)+g_{x^2-y^2}^{\sigma}(E)+g_{xy}^{\sigma}(E)]\}dE, \tag{A2}$$

and for an arbitrary angle ϑ with respect to the z axis,

$$M_L^{00}(\vartheta)=M_L^{00}(0)+\Delta M_L^{00}\sin^2\vartheta, \tag{A3}$$

with $\Delta M_L^{00}=M_L^{00}(\pi/2)-M_L^{00}(0)$. For the systems with O_h symmetry $g_{xy}^{\sigma}=g_{xz}^{\sigma}=g_{yz}^{\sigma}\equiv g_{t_{2g}}^{\sigma}$ and $g_{x^2-y^2}^{\sigma}=g_{z^2}^{\sigma}\equiv g_{e_g}^{\sigma}$. Thus

$$M_L^{00}=\xi\sum_{\sigma=\pm 1}\sigma\int_{-\infty}^{E_F}\mathrm{Im}\{g_{t_{2g}}^{\sigma}(E)[4g_{e_g}^{\sigma}(E)+g_{t_{2g}}^{\sigma}(E)]\}dE \tag{A4}$$

and its magnitude does not depend on orientation $[M_L^{00}(0)=M_L^{00}(\pi/2)]$.

APPENDIX B: ORBITAL POLARIZATIONEFFECTS AND MAE

In order to include the orbital polarization (OP) effects in the MAE calculations let us consider the model Hartree-Fock (HF) approach. We do not make any restrictions on the form of the orbitals used in the HF approximation and write the energy for on-site electron-electron interaction in general form:

$$E_{ee}^{i}=\frac{1}{2}\sum_{\alpha\beta\gamma\delta}[U(\alpha\gamma\beta\delta)-U(\alpha\gamma\delta\beta)]n_{\alpha\beta}^{i}n_{\gamma\delta}^{i}, \tag{B1}$$

where the greek indices correspond to the pairs $(m\sigma)$ of azimuthal and spin quantum numbers, $\|n_{\alpha\beta}^{i}\|$ is the matrix of occupations on the ith site related to the Green's function as

$$n_{\alpha\beta}^{i}=-\frac{1}{\pi}\mathrm{Im}\int_{-\infty}^{E_F}dEG_{\alpha\beta}^{ii}(E), \tag{B2}$$

$U(\alpha\gamma\beta\delta)=\langle\alpha\gamma|1/r_{12}|\beta\delta\rangle$ are four-index matrices of Coulomb interaction which can be expressed through Clebsch-Gordan coefficients and renormalized Slater's integrals.

Now let us consider the SOI as a small perturbation and present $n_{\alpha\beta}$ in the form $n_{\alpha\beta}^{i}=n_{\alpha\beta}^{0i}+\delta n_{\alpha\beta}^{i}$, where $n_{\alpha\beta}^{0i}$ corresponds to the scalar-relativistic case and $\delta n_{\alpha\beta}^{i}$ is a correction of first order in ξ:

$$\delta n_{\alpha\beta}^{i}(\vartheta)=-\frac{1}{\pi}\sum_{j}\mathrm{Im}\int_{-\infty}^{E_F}dE(\tilde{G}_0^{ij}\Delta V^{j}\tilde{G}_0^{ji})_{\alpha\beta}. \tag{B3}$$

Then, using arguments analogous to the local-force theorem,[38] we find that the change of the total energy due to the SOI can be presented as a change of the one-electron part plus the electron-electron interaction correction

$$\delta E_{ee}^{i}(\vartheta)=\frac{1}{2}\sum_{\alpha\beta\gamma\delta}[U(\alpha\gamma\beta\delta) - U(\alpha\gamma\delta\beta)]\delta n_{\alpha\beta}^{i}(\vartheta).\delta n_{\gamma\delta}^{i}(\vartheta). \tag{B4}$$

Due to symmetry properties of the SOI we have $\Sigma\alpha\in\{\sigma\}\delta n_{\alpha\alpha}^{i}=0$ for every projection of spin σ. Thus, similar to the single-particle energies, contributions of the first-order corrections $\delta n_{\alpha\beta}^{i}$ to the spin density vanish and the effect described by Eqs. (B3) and (B4) is essentially beyond the local-spin-density approximations. Together with the change of the one-electron energy, the electron-electron interaction correction is an effect of second order with respect to the spin-orbit coupling (we note that in LSDA the corresponding correction of the double-counting term appears only in fourth order ξ).

We estimated the $\delta E_{ee}(\vartheta)$ on every site of considered compounds using the single-impurity approximation for the SOI. Then, the renormalized parameters of electron-electron interaction can be found using a constraint-LSDA scheme (see Ref. 39 for details). Numerically we found that for the uniaxial crystals $\delta E_{ee}(\vartheta)$ also exhibit the sine-square dependence on the orientation of magnetization with respect to the perpendicular axis. The OP to the MAE is negative for CoPt (correspondingly -0.011 and -0.009 mRy on Co and Pt sites). Thus, it could improve agreement with experimental data for this compound, whereas for FePt, where the OP effect obtained in the single-impurity approximation is positive, the discrepancies become even worse. A proper consideration of the OP effects in the context of MAE calculations taking into account more appropriate spatial extension beyond the single-site approximation is evidently needed.

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