

Magnetic order and anisotropy in hexagonal MnPtGa: Noncollinear magnetism contra frustration

Gerhard H. Fecher^{✉,*}, Roshnee Sahoo^{✉,†} and Claudia Felser^{✉,‡}
Max-Planck-Institute for Chemical Physics of Solids, D-01187 Dresden, Germany



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MnPtGa is a hexagonal intermetallic compound with a rich variety of magnetic orders. Its magnetic state is reported to range from collinear ferromagnetism, to noncollinear skyrmion-type order. MnPtGa is a system with strongly localized magnetic moments at the Mn atoms, as was demonstrated using calculations for disordered local moments. The magnetic moments at the Mn sites stay at $\approx 3.9 \mu_B$ even above the calculated magnetic transition temperatures ($T_N = 220$ K or $T_C = 285$ K). In the present work, a special emphasis was placed on the possible noncollinear magnetic order using first-principles calculations. The investigations included magnetic anisotropy, static noncollinear orders in the form of spin canting, and dynamic noncollinearity in spin spirals. It is found that the energy differences between ferromagnetic, antiferromagnetic, canted, or spiral magnetic order are in the order of not more than 30 meV, which is in the order of thermal energies at ambient temperature. This hints that a particular magnetic state—including skyrmions, antiskyrmions, or spin glass transitions—may be forced when an external field is applied at finite temperature.

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I. INTRODUCTION

Hexagonal, magnetic ternaries based on d elements are interesting as they might exhibit noncollinear spin structures, as observed, for example, in the hexagonal rare earth elements. Antiferromagnetism in the basal plane might lead to frustration due to the triangular arrangement of the atoms. As a result, spin glass type, spiral spin order, or other noncollinear magnetic order may appear.

MnPtGa, with a hexagonal crystal structure, was selected for the present study. In an early report, it was unfortunately classified as a Heusler compound with a cubic $C1_b$ structure [1]. Buschow and co-workers [2,3], however, have shown that MnPtGa adopts a hexagonal Ni_2In derived crystal structure with lattice parameters $a = 4.328$ Å and $c = 5.576$ Å. Ferromagnetic order was reported with a magnetic moment of 3.15 to $4.72 \mu_B$ per manganese atom and a Curie temperature in the order of 210 to 270 K [2–4]. These early studies were all performed for temperatures above 200 K; thus, the low-temperature magnetic structure stayed rather unexplained. Cooley *et al.* [5] reported a variety of different types of collinear and noncollinear magnetic orders while varying the temperature between 4 and 300 K. The series of different

magnetic order reported in Ref. [5] is: collinear ferromagnetic at 200 K, canted ferromagnet at 150 K, canted ferromagnet with spin density wave at below 50 K with change of the period, and increased intensity by further cooling to 10 K. The pressure dependence of the magnetic order was discussed by Dubey *et al.* [6] for polycrystalline MnPtGa. Recently, measurements on single-crystalline MnPtGa were reported and some aspects of the influence of the noncollinear magnetic order on the band structure were explained by Dwari *et al.* [7].

Besides the above-mentioned noncollinear behavior, skyrmions were observed in the Mn–Pt–Ga system at temperatures below 220 K [8]. It was reported that the stability of the Néel skyrmions depends on the thickness of the investigated lamella. This was observed over a wide range of temperatures. Further, a noncollinear magnetic order, with a canting of the magnetic moments similar to the bulk material, was detected in epitaxial MnPtGa thin films [9,10]. Non-orthogonal spin currents were recently investigated in MnPtGa films grown on MgO by Meng *et al.* [11].

In the present work, collinear and noncollinear magnetic properties of MnPtGa are investigated by means of first-principles methods. Ferromagnetic and antiferromagnetic interactions are investigated, as well as properties related to canted or spiral spin order.

II. DETAILS OF THE CALCULATIONS

The electronic and magnetic structures of MnPtGa were calculated by means of WIEN2k [12–14] and SPRKKR [15–18] in the generalized gradient approximation (GGA). In particular, the GGA functional in the parametrization of Perdew, Burke, and Ernzerhof [19] was used. A test involving GGA + U resulted in magnetic moments that were too high compared with experimental results. WIEN2k was used for the structural

*Contact author: fecher@cpfs.mpg.de

†Present address: Binayak Acharya Degree College, Berhampur 760006, Odisha, India.

‡Contact author: felser@cpfs.mpg.de

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relaxation (volume and c/a) and to test the magnetocrystalline anisotropy. (Note that the forces on the atoms are identical to zero, due to the symmetry of their positions. Therefore, their relative positions are not changed during relaxation of the structure.) All other properties were calculated using SPRKKR [16]. Magnetic spin and orbital moments, magnetic anisotropy, exchange coupling coefficients, Dzyaloshinskii–Moriya vectors, transition temperatures, disordered local moments, and canted spin order were all determined in the fully relativistic mode, solving the spin-polarized Dirac equation for core and valence electrons.

The spin-polarized semirelativistic mode (Dirac equation only for core electrons) was used for spin spirals, due to symmetry constraints [20,21]. To visualize the electronic structure, the Bloch spectral functions ($E(k, \rho)$) [22] were calculated using the fully relativistic mode. $E(k, \rho)$ can be interpreted as a kind of momentum (k) resolved density of states (ρ). It is similar to the band structure. The mean-field approach of Liechtenstein was used to determine the magnetic transition temperatures [23]. For multisublattice systems, including antiferromagnets, Anderson’s approach is used [24]. Due to the long-range interaction and the slow convergence of the exchange coupling coefficients with cluster size, the spin wave stiffness constants and the spiralization coefficients can be determined using the extrapolation scheme of Pajda *et al.* [25,26]. More details of the calculations are found in Refs. [27,28], as well as in additional citations given in the following sections.

The basic crystal structure of MnPtGa (prototype: BeZrSi; $hP6$; $P6_3/mmc$ (194) *cad*) is shown in Fig. 1 for different magnetic structures. The atoms are placed in the paramagnetic or ferromagnetic structure on the 2a, 2c, and 2d Wyckoff positions of the hexagonal cell. For the calculation of the antiferromagnetic structure, the Mn position is split, the lattice symmetry is reduced to $P\bar{3}m1$ (164) where the $Mn_{\uparrow}$, $Mn_{\downarrow}$, and Pt, Ga atoms are placed on 1a, 1b, and 2d (with different position parameters for Pt and Ga), respectively.

The magnetic order changes the symmetry [29]. Depending on the direction of magnetization, incompatible symmetry elements must be changed or removed. This results in the magnetic (colored) space groups listed in Ref. [30]. The magnetic symmetry is not necessarily the same as the lattice symmetry, but it is generally lower. The magnetic space groups for various directions of magnetization can be determined using FINDSYM [31,32]. The magnetic space group is $P6_3/mmc'$ (194.270) for the collinear ferromagnetic order with spin magnetic moments m_s along the c axis. For the antiferromagnetic order, it becomes $P6_3'/m'mc$ (194.268) when $m_s \parallel c$. Here, the prime ' is the spin reversal operator [29] (sometimes called the time reversal operator). m' is a mirror operation followed by spin reversal. It should be mentioned that the colored group is $Cm'c'm$ (63.462) or $Cmcm$ (63.457) for the antiferromagnetic order with magnetic moments lying in the basal plane. However, a more complicated checkerboard-type antiferromagnetic order may also appear. For the later investigated noncollinear spin structure, the magnetic space group becomes $P2_1/m.1$ (11.50) or $Cm'c'm$ (63.462). In the simple antiferromagnetic or noncollinear cases, the Wyckoff positions are different, but the lattice parameters are kept hexagonal.

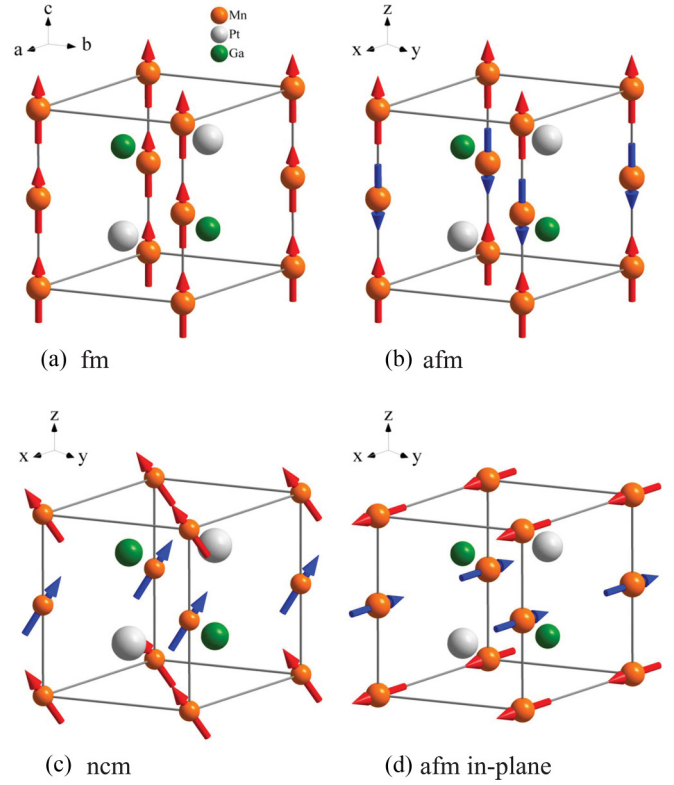


FIG. 1. Crystal and magnetic structures of MnPtGa. (a) ferromagnetic, (b) antiferromagnetic, (c) noncollinear, (d) antiferromagnetic in-plane. The arrows are drawn in the direction of the local magnetic moments at the Mn atoms. The angle between the magnetic moment vectors and the z axis is in the canted, noncollinear state (c) is $\theta = 35^\circ$, which is close to $\arctan(a/c) \approx 38^\circ$.

III. RESULTS AND DISCUSSION

A. Collinear magnetic structures

The lattice parameters reported from experiments vary and are in the ranges ($4.328 \leq a \leq 4.446$) Å and ($5.572 < c \leq 5.590$) Å [2–5,8]. Therefore, calculations of the electronic structure and magnetic properties were performed for optimized lattice parameters. As a starting point, the lattice parameters of the two collinear magnetic structures were optimized using WIEN2k. Spin-orbit interaction was accounted for to respect the high Z of Pt and the spin-orbit splitting of the Ga 3d semicore level. The results of the optimization for collinear spin structures are summarized in Table I. The calculated lattice parameters and positions of the atoms fit very well to those reported from experiments [2–5].

The equilibrium lattice parameters of the ferromagnetic and antiferromagnetic states are very close together. For a better comparison, the Birch–Murnaghan [33,34] equation of state energy versus volume and pressure curves for the two collinear magnetic orderings are shown in Fig. 2. The equilibrium energies at ambient pressure ($p = 0$) differ by only 33 meV, that is, in the order of thermal energies at room temperature. The lowest energy is obtained for the antiferromagnetic state (see also Table I). This gives a hint that the low-temperature magnetic state may differ from the high-temperature ferromagnetic state observed in experiments.

TABLE I. Properties of MnPtGa with ferromagnetic (fm) or antiferromagnetic (afm), collinear magnetic order. Tabulated are the lattice parameters (a , c , c/a), the Bulk modulus B for hydrostatic pressure (fixed optimized c/a ratio), the total energy (E_{tot}), and the magnetic moment at the Mn atom m_{Mn} from the optimization with WIEN2k. Experimental lattice parameters are from Refs. [2–5,8]. Experimental magnetic moments from [2,5] depend on the temperature.

	fm	afm	experiment
a [Å]	4.3552	4.3571	4.328 ... 4.446
c [Å]	5.5306	5.5318	5.572 ... 5.590
c/a	1.2699	1.2696	1.257 ... 1.289
B [GPa]	112	151	
E_{tot} [Ry]	−86198.140678	−86198.143094	
m_{Mn} [μ_B]	3.7232	±3.7105	2.63 ... 3.15

The magnetic moment at the Mn atoms is nearly independent of the magnetic order. The small difference is caused by the slightly different lattice parameters. Its value of $\approx 3.7 \mu_B$ as well as the ferromagnetic Curie temperature are in agreement with the findings from experiments [2–4]. The calculated Curie temperature is 65 K higher compared to the Néel temperature. Therefore, it can be concluded that the high- and low-temperature magnetic order is different, as mentioned above.

The exchange coupling coefficients J_{ij} needed to calculate the transition temperatures [23,25,35,36] are plotted in Fig. 3 for the case of ferromagnetic or antiferromagnetic ordered collinear magnetic moments. In micromagnetic or atomistic spin dynamics calculations, magnetic order is ferromagnetic when the intersublattice exchange coupling coefficients are positive, and antiferromagnetic when they are negative. In electronic structure calculations, the individual J_{ij} values can

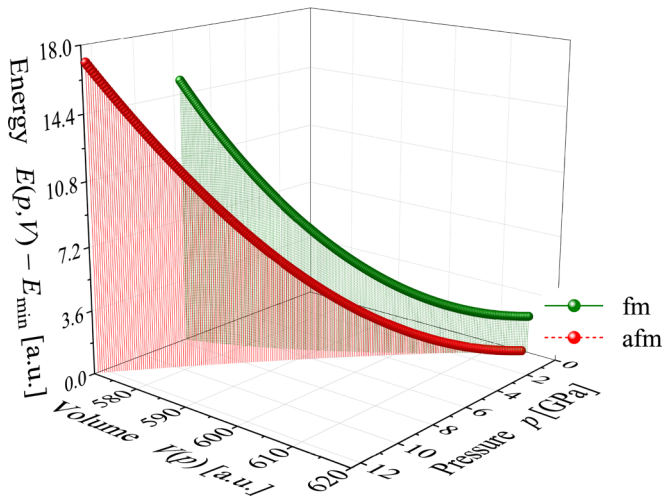


FIG. 2. Calculated equation of state for MnPtGa with ferromagnetic (fm) or antiferromagnetic (afm), collinear magnetic order. Shown are the energy–volume–pressure relations ($E(V, p) - E_{\text{min}}$) according to the Birch–Murnaghan equation of state [33,34]. E_{min} is the equilibrium energy of the antiferromagnetic state (see Table I). Magnetization is along [0001]. [Note the use of atomic units for energy (mRy) and volume (a_{0B}^3 , $a_{0B} = 0.529 \dots \text{Å}$).]

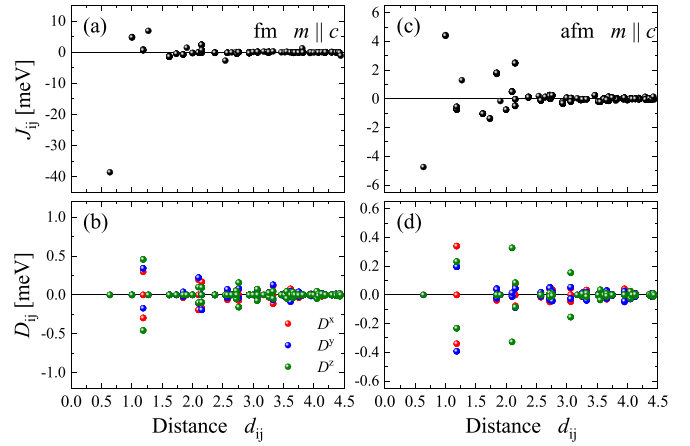


FIG. 3. Exchange coupling coefficients of MnPtGa with ferromagnetic (fm) or antiferromagnetic (afm) order. Compared are the exchange coupling coefficients J_{ij} in (a),(c), and the Dzyaloshinskii–Moriya coefficients D_{ij} in (b),(d). The distance $d_{ij} = |r_{ij}|/a$ between the Mn atoms is given relative to the lattice parameter a . Magnetic moments are assumed to be aligned along the c axis. (Note the different energy scales.)

be either positive or negative depending on the distance r_{ij} , regardless of the magnetic order. The large negative values for the interaction between nearest neighbors indicate that the ferromagnetic state is not stable, at least at low temperatures. Further shown are the antisymmetric exchange constants $D_{ij}^{x,y,z}$ [35] that are the components of the Dzyaloshinskii–Moriya interaction [37,38] vectors. The D_{ij} are considerably smaller than the J_{ij} ; however, the latter drops down faster with increasing distance between the Mn atoms.

The isotropic J_{ij} and anisotropic D_{ij} exchange coupling coefficients form the exchange tensor:

$$\mathbb{J}_{ij} = \begin{pmatrix} J_{ij} & D_{ij}^z & -D_{ij}^y \\ -D_{ij}^z & J_{ij} & D_{ij}^x \\ D_{ij}^y & -D_{ij}^x & J_{ij} \end{pmatrix}, \quad (1)$$

where $J_{ij} = \frac{1}{3} \sum_{\alpha} J_{ij}^{\alpha\alpha}$ and $D_{ij}^{\gamma} = \frac{1}{2} (J_{ij}^{\alpha\beta} - J_{ij}^{\beta\alpha})$ ($\alpha, \beta, \gamma = x, y, z, \alpha \neq \beta \neq \gamma$) [39,40].

Further, the exchange coupling coefficients may be used to determine effective spin model parameters for micromagnetic calculations. These are the spin wave stiffness (A) and spiralization (D):

$$A = \sum_{i \neq j} J_{ij} (r_{ij} \otimes r_{ij})$$

$$D = \frac{1}{2} \sum_{i \neq j} D_{ij} \otimes r_{ij}, \quad (2)$$

where r_{ij} are the difference vectors between the positions of atoms i and j ($i \neq j$) and $\otimes$ is the tensor product [40,41]. The spin stiffness constant A describes a quadratic dependence of the spin wave energy on the wave vector q , that is, $E_{\text{SW}} = Aq^2$. It should not be confused with the exchange stiffness constant.

The allowed combination of the vector components D_{ij}^{α} of the exchange coupling coefficients and spatial coordinates r_{ij}^{β}

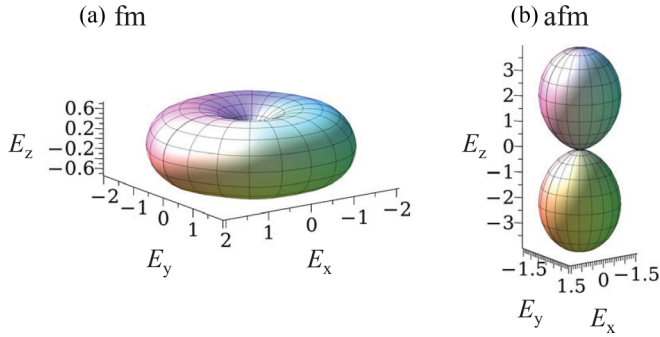


FIG. 4. Magnetic anisotropy of ferromagnetic (fm) and antiferromagnetic (afm) MnPtGa. The magnetocrystalline anisotropy energy E_a' is given in meV. In the ferromagnetic state, the “easy” direction is along z ([0001]), whereas three principal “hard” directions appear in the basal plane. The basal plane becomes an “easy” plane in the antiferromagnetic state.

depends on the investigated geometry (α, β assign the vector components x, y, z of D_{ij} and r_{ij}).

The general components of the spiralization matrix are $D^\nu = \sum_{i \neq j} D_{ij}^\alpha r_{ij}^\beta$. For a magnetic monolayer (ML) with C_{3v} ($m\bar{3}$) point-group symmetry, the effective stiffness and spiralization are $A_{ML} = \frac{1}{4} \sum J_{ij} (d_{ij}^y)^2$ and $D_{ML} = \sum D_{ij}^x \cdot d_{ij}^y$, respectively, [42] (here, $d_{ij} = r_{ij}/a$).

All D_{eff} are zero in the present case because the investigated bulk structure shown in Fig. 1 is centrosymmetric [37]. The Mn sites have D_{3d} ($\bar{3}m$) point group symmetry. It is usually expected that skyrmions require noncentrosymmetric crystal structures in which the spiralization does not vanish. However, here the bulk material is centrosymmetric, which would render the observed appearance of skyrmions impossible. At the surface, however, the inversion symmetry is broken [43] because an inversion would equate the bulk to the vacuum. Similarly, inversion symmetry is broken at the interface with a different material. This suggests that skyrmions could exist in thin films despite the centrosymmetry of the bulk material. Due to geometric restrictions in the summation, in thin films, the spiralization may not vanish. This is analogous to the dipole anisotropy, which can differ between bulk materials and thin films.

Thus, the vanishing of the spiralization for bulk materials does not contradict the observed existence of skyrmions. In thin films or lamellas, the summations in the effective stiffness and spiralization are spatially restricted in at least one direction, which can lead to a nonvanishing D_{eff} . This is a direct consequence of broken inversion symmetry at surfaces and interfaces. This possibly explains the experimental observation by Srivastava *et al.* [8] that the size and stability of the skyrmions depend on the thickness of the investigated lamella.

Further, the directional dependence of the magnetization was investigated to explain details of the collinear magnetic order. In particular, the total energy was calculated for the cases where the magnetization is pointing along different crystallographic directions. The obtained energy differences allow one to determine the magnetocrystalline anisotropy (see also Appendix A). The result is visualized in Fig. 4.

In the ferromagnetic state, the anisotropy is almost uniaxial with the “easy” axis (lowest energy) along z ([0001]), and an appreciable variation in the basal plane. It is hardest along x , that is, in the direction between the nearest neighbors in the antiferromagnetic state. Here, the z axis and is the “hard” axis (highest energy) pointing along [0001]. The basal plane becomes an easy plane. A closer inspection reveals that there are three main easy axes found in the basal plane that are along x ($[21\bar{3}0]$) and similar. However, the energy differences to the y axis ($[01\bar{1}0]$) are very tiny only; thus, that already small changes in the lattice parameters, method, and parameters of the calculations are able to change the situation (see also Ref. [44] for a comparison of different methods). The second-order uniaxial anisotropy constants are $K_u^{\text{fm}} = 2.85$ MJ/m³ and $K_u^{\text{afm}} = -6.85$ MJ/m³ for the ferromagnetic and antiferromagnetic states, respectively. These values result in anisotropy fields of $\mu_0 H_a^{\text{fm}} = 7.5$ T and $\mu_0 H_a^{\text{afm}} = 18.1$ T. From these observations, it is concluded that the ferromagnetic order prefers a nearly uniaxial anisotropy with c an easy axis, whereas the result of antiferromagnetic order is a triaxial anisotropy with the spins aligned in the basal plane. In both cases a and b are hard axes. Care has to be taken if the anisotropy energy is determined while applying a magnetic field which changes the magnetic energy. The consequences are examined in Appendix B.

Further, the dipolar magnetocrystalline anisotropy was calculated from a direct lattice sum of the dipolar energy (see Appendix C):

$$E_{\text{dipanis}} = E_{\text{dip}}(\vec{n}_1) - E_{\text{dip}}(\vec{n}_2), \quad (3)$$

where $\vec{n} = \vec{M}/M$ is the direction of magnetization. Here, the anisotropy energy $\Delta E_{\text{dipanis}}$ is the difference of the dipolar energies for the two different directions $\vec{n}_1 = [0001]$ and $\vec{n}_2 = [1000]$. These are the directions along the c axis and in the basal plane along a . Using these directions in Eq. (3), the dipolar magnetic anisotropies for the collinear, ferromagnetic, and antiferromagnetic states are found to be $\Delta E_{\text{dipanis}}^{\text{fm}} = -74$ μeV (-0.13 MJ/m³) and $\Delta E_{\text{dipanis}}^{\text{afm}} = 119$ μeV (0.21 MJ/m³), respectively. These values are about 20 to 30 times lower compared to the magnetocrystalline anisotropy energies. The positive value of the antiferromagnetic state points in an easy dipolar direction that is in the basal plane, whereas the ferromagnetic state has an easy dipolar axis along the c axis.

The dipolar anisotropy is rather small compared to the anisotropy calculated from the total energies. Here, it was calculated for a sphere with a radius of 50 nm. The situation will change for other shapes, resulting in a distinct shape anisotropy. In particular, in thin films, the dimension perpendicular to the film is much smaller compared to the dimensions in the film plane. Therefore, the summation in Eq. (C1) of Appendix C becomes a truncated sphere that is strongly anisotropic, and one will obtain a pronounced thin film anisotropy. Additionally, this thin film anisotropy will be affected by magnetic moments that differ at interfaces and surfaces from the center of the film.

TABLE II. Magnetic properties of MnPtGa. Tabulated are the total energies (E_{tot}), the magnetic moments at the Mn atoms (m_{Mn}), the uniaxial crystalline anisotropies (K_u), the dipolar anisotropies ($\Delta E_{\text{dipanis}}$), and the magnetic transition temperature T_m , where $m = C$, or N for the Curie or Néel temperature, respectively. Calculations were performed with SPRKKR. Experimental T_m values are for ferromagnetic transitions [2–5,9,10]. The magnetic moments from [2,5] depend on the temperature.

	fm	afm	ncm	experiment
$ m_{\text{Mn}} [\mu_B]$	3.72	3.71	3.18	2.63 ... 3.15
T_m [K]	285	220		210 ... 263
K_u [meV]	1.61	−3.89		
$\Delta E_{\text{dipanis}}$ [meV]	−0.07	0.12		

The results of the calculations with SPRKKR for the magnetic properties of MnPtGa in the collinear state are summarized in Table II.

B. Disordered local moments

Compounds containing Mn are typically systems with localized magnetic moments. Here, the coherent potential approximation (CPA) [45,46] was used to model the random distribution of the magnetic moments at the Mn atoms, which is known as the disordered local moment (DLM) [47,48] method. At each Mn site, two species with opposite directions and equal sizes of the magnetic moments were assumed, starting from 99% spin up ($\uparrow$) and 1% spin down ($\downarrow$) up to 50% of both spin directions. That is, the calculations were performed for the pseudo-alloy $(\text{Mn}_{(1-x)\uparrow}\text{Mn}_{x\downarrow})\text{PtGa}$. It was found that the magnetic moment at the Mn sites stays in the order of $3.9 \mu_B$ whereas the total magnetic moment of MnPtGa vanishes (see Fig. 5). This is in good agreement with the experimental

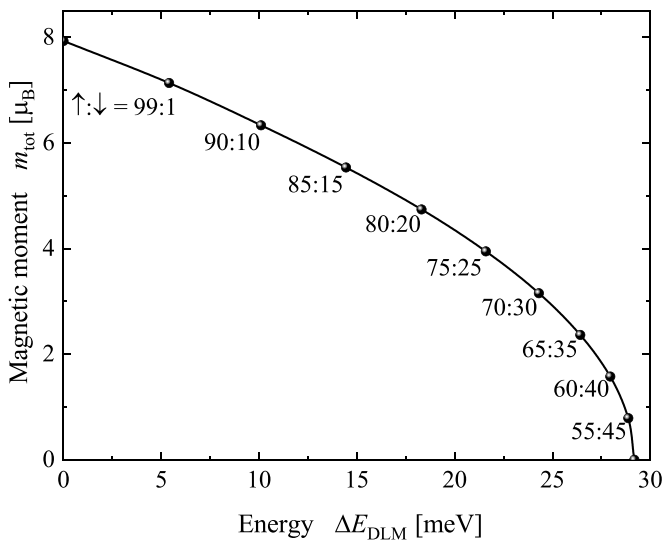


FIG. 5. Disordered local moments in MnPtGa. The total (spin plus orbital) magnetic moment in the primitive cell containing 2 Mn atoms is plotted as a function of the energy difference between the ferromagnetic and disordered local moment state. $\uparrow\downarrow$ is the ratio of spin up to spin down electrons at the Mn site.

work of Buschow and de Mooij [3], who found an effective paramagnetic moment of $3.6 \mu_B$ per Mn atom from the slope of the inverse susceptibility $\chi^{-1}(T)$ curve at high temperature ($T > 300$ K).

Figure 5 shows the total magnetic moments (spin + orbital) in the primitive cell (containing two Mn atoms) as a function of the energy difference between the fully ferromagnetic and completely spin-disordered states. The energy difference for complete spin disorder amounts to 29.2 meV. This corresponds to $\Delta E/k_B \approx 170$ K per Mn atom.

Figure 6 compares the electronic structure of MnPtGa with ordered and disordered local magnetic moments. In the disordered local moment state, the electronic states in the region of the Fermi energy (ϵ_F) are strongly broadened compared to the case with ordered moments, whereas those farther away from ϵ_F are less affected. Compared to the ferromagnetic state, it is found that the exchange splitting is smaller in the disordered local moment state. This becomes obvious for the band minima at the M point where the splitting finally vanishes for full spin disorder or in the antiferromagnetic state. It is not possible to directly compare Fig. 6 to the experiments of Cooley *et al.* [5], because the magnetic order changes with temperature and cannot directly be compared to the calculations for the purely collinear ferromagnetic state. Furthermore, these types of calculations cannot be performed with an applied magnetic field, which is used in the experiments.

C. Spiral spin order

The close energies and phase transition temperatures for the ferromagnetic and antiferromagnetic states, together with the more complicated behavior of the antiferromagnetic anisotropy, suggest that a noncollinear magnetic order appears in MnPtGa. Therefore, spin spirals (see Refs. [20,21,49] for details) were calculated for different directions and different starting magnetic spin orders, either antiferromagnetic or ferromagnetic. In centrosymmetric systems, the spin spirals are not influenced by spin-orbit or Dzyaloshinskii–Moriya interaction [37]. Note that the orientation of the spins will no longer be the same as shown in Fig. 1, but they will depend on the direction of propagation as well as the initial magnetic order. In planar spirals, the spins are perpendicular to the propagation direction. The energies are compared in Fig. 7 for such planar-type spirals.

The spirals along x or y propagate in the hexagonal planes, where y is in the direction of a nearest neighbor atom (note: here, y corresponds to $[01\bar{1}0]$ in the hexagonal crystal coordinates). In these cases, the lowest energy is observed at $q = 0$ independent of the initial spin order. Most interesting is the behavior along the z axis ($[0001]$). For both initial spin orders, the energy is clearly lower for $q = \pi/d_{0001}$ compared to 0. The energy differences amount to about -20 meV. The energy is nearly constant in the antiferromagnetic spiral up to $\approx 0.5\pi/d_{0001}$. Similarly, it is observed in the ferromagnetic spiral that the energy is nearly constant above $\approx 0.5\pi/d_{0001}$. Both might hint at frustrated states, as many spin configurations will all have the same energy. Further, when starting with ferromagnetic spin order, the energy exhibits a local minimum at $\approx 0.5\pi/d_{0001}$. A discussion of the conditions for transitions

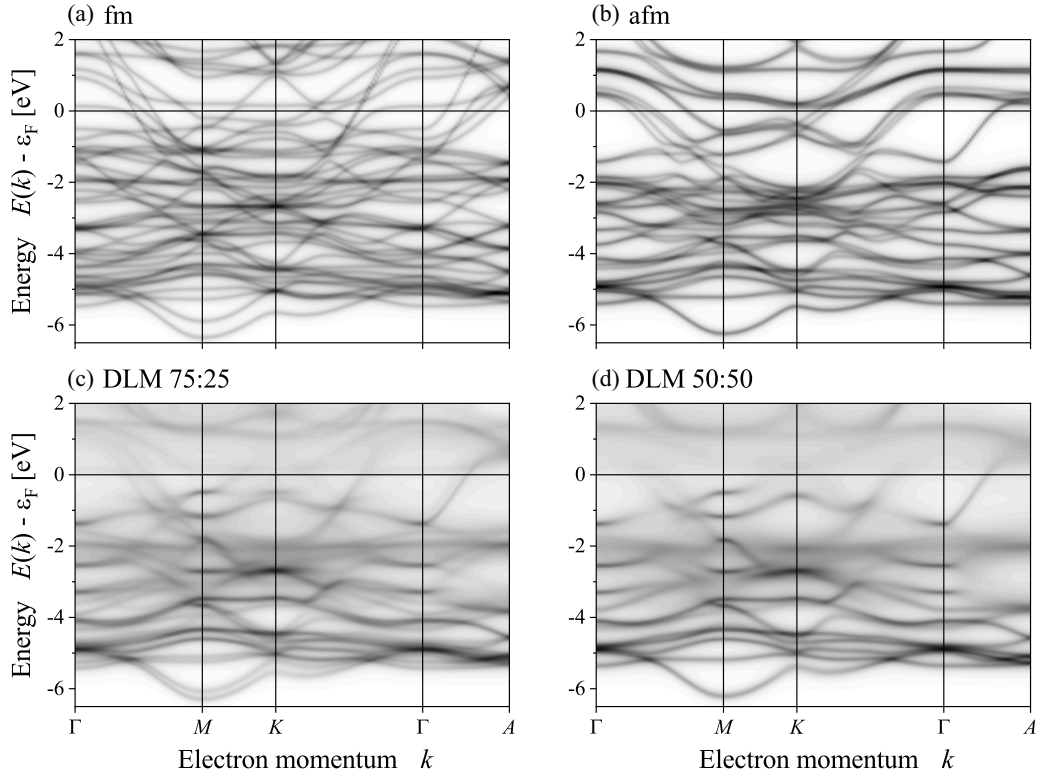


FIG. 6. Electronic structure for ordered and disordered local moments in MnPtGa. The Bloch spectral functions of the ferromagnetic (a) and antiferromagnetic (b) states are compared to the disordered local moment states with 50% (c) and full (d) spin disorder.

between collinear (e.g., ferromagnet) or noncollinear (e.g., spin spiral) arrangements of the magnetic moments in elements was provided by Lizárraga *et al.* [50]. Planar spin

spirals were also used to explain the magnetic frustration, the spiral spin order, as well as the multidegenerate ground state of the rubidium sesquioxide Rb_4O_6 [51].

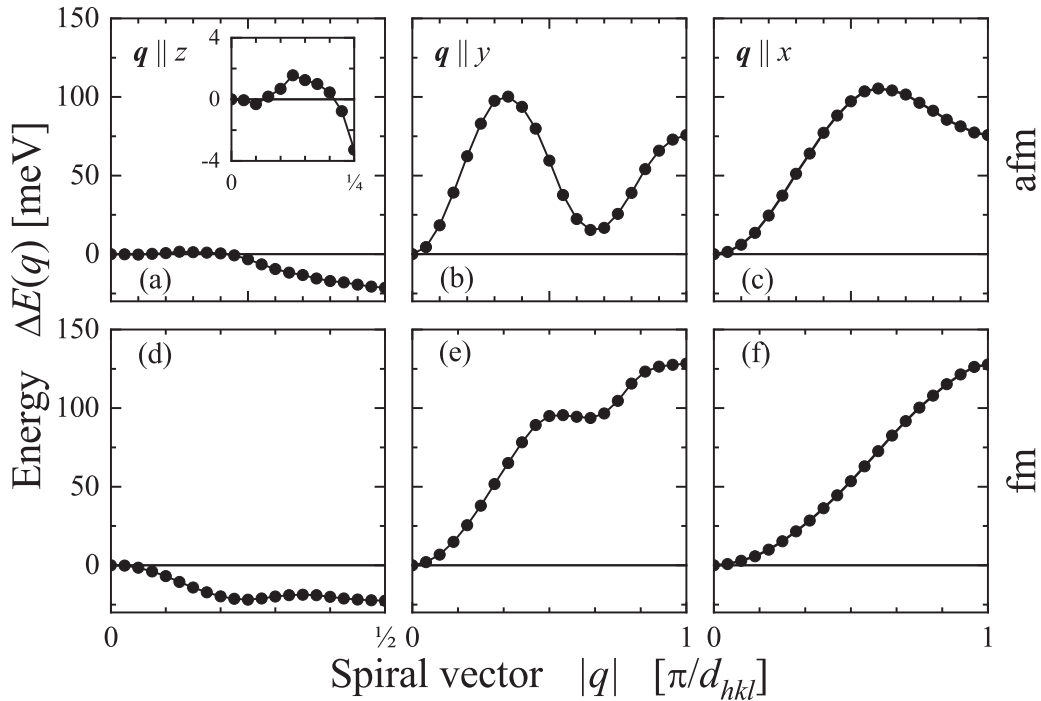


FIG. 7. Planar spin spirals in antiferromagnetic (a)–(c) and ferromagnetic (d)–(f) MnPtGa. The spiral energies are given with respect to $q = 0$, that is: $\Delta E(q) = E(q) - E(0)$. Note that the absolute energies at $q = 0$ differ between the starting spin orders but are identical for different directions within a given type of magnetic order.

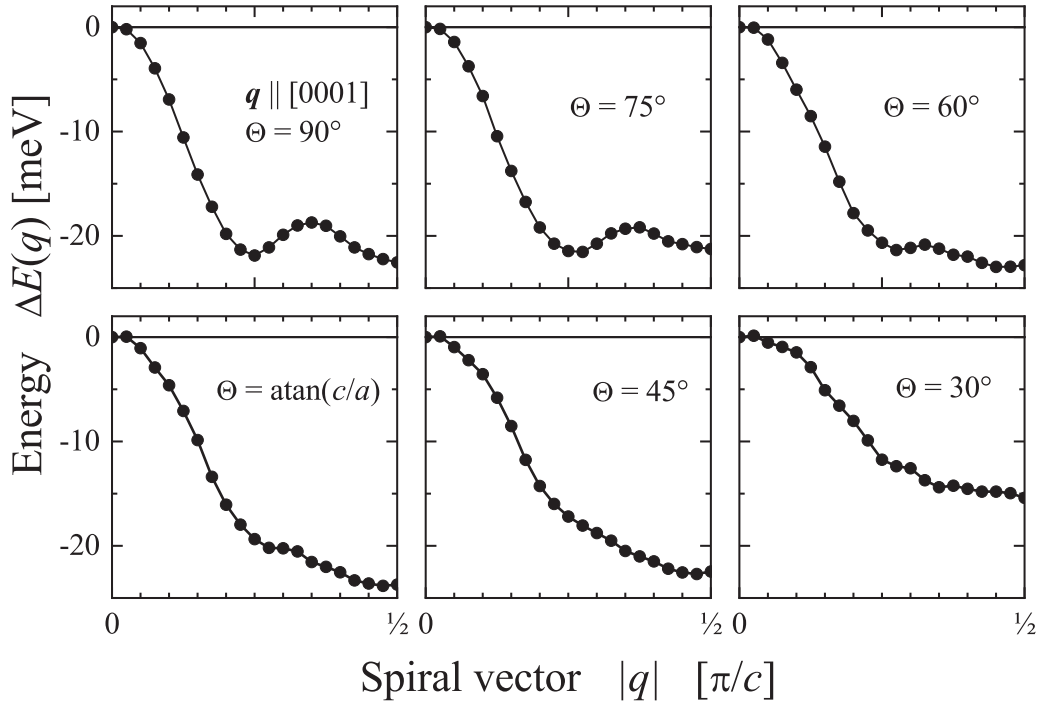


FIG. 8. Energies of ferromagnetic cone spirals.

The spin direction was assumed to be perpendicular to the q vector, in the above calculations for planar spirals. Thus, the angle between $\vec{q}$ and local magnetic moments $\vec{m}_i$ was set to $\Theta = \pi/2$. In the next step, the spirals were assumed to be of cone type with $0 < \Theta < \pi/2$, to allow for a more detailed analysis. The calculations were performed for q along [0001]. The energies for q in other directions are, of course, higher throughout, as already observed for the planar spirals. Figure 8 displays the results for the ferromagnetic cone spirals with variation of the cone angle. The lowest energy always appears at $q = 0.5\pi/c$. The local energy minimum observed in the planar spiral vanishes with decreasing cone angle. The ferromagnetic, cone-type spin spirals have a negative curvature at $q = 0$, regardless of the propagation direction. Therefore, these spiral structures exhibit negative spin stiffness constants. This corresponds to the previously observed negative values of the exchange coupling coefficients between nearest neighbors for collinear spins.

The results for the antiferromagnetic cone spirals are displayed in Fig. 9. Similar to the ferromagnetic case, the lowest energy always appears at $q = 0.5\pi/c$. Starting from the planar spiral with $\Theta = \pi/2$, the q range with nearly unchanged energies increases slightly.

The dependence of the energy minimum on the cone angle Θ is compared in Fig. 10. The minimum energy of the antiferromagnetic cone spiral decreases with decreasing cone angle. It is lowest for $\Theta = \pi/2$, that is, for the planar spiral. The behavior of the ferromagnetic cone spiral is different. After an initial decrease, only a weak variation is observed for cone angles above 45° , and the lowest value appears already for $\tan(\Theta) \approx c/a$ corresponding to approximately 51.78° . As a result, one has a spiral with a ferromagnetic moment along its propagation direction. The small energy variations at large Θ

as well as large q again hint at frustration, in particular in the basal plane of the lattice.

D. Canted spin order

In a last step, the energies were calculated for noncollinear, canted spin orientation. It was assumed that the magnetic moments are aligned under a polar angle θ with respect to the c axis. The azimuthal angle ϕ is measured with respect to the x axis (corresponding to $[21\bar{3}0]$ in crystal co-ordinates). The starting point is the antiferromagnetic structure with two distinguished Mn atoms. The magnetic moments at the two Mn atoms were set to have a ferromagnetic (parallel) orientation along the c axis and an antiferromagnetic (antiparallel) alignment in the basal plane by setting $\phi_2 = \phi_1 + \pi$. It turned out that the lowest energies are obtained for $\phi_{1,2} = 0, \pi$. The resulting energies and magnetic moments are shown in Fig. 11.

The ferromagnetic contribution to the spin magnetic moment varies between $\theta = 0$ and $\pi/2$ from $3.81 \mu_B$ per primitive cell to 0, while going from a ferromagnetic to an antiferromagnetic state. At the same time, the energy exhibits a minimum at $\theta \approx 38^\circ$, that is, about 25 or 30 meV lower compared to that of the antiferromagnetic or ferromagnetic states, respectively. From this observation, it is concluded that the magnetic ground state of MnPtGa is most probably canted. The corresponding magnetocrystalline structure is shown in Fig. 1(c). [Note that the angle from Mn to a second nearest neighbor Pt atom is 31.2° , and one finds, for example, that the magnetic moment is approximately pointing from the Mn at $(0, 1, 0)$ to the Pt atom at $(1/3, 2/3, 3/4)$.] The canting angle at the minimum of energy calculated here for bulk material is larger compared to the one of $\approx 20^\circ$ determined at 2 K for

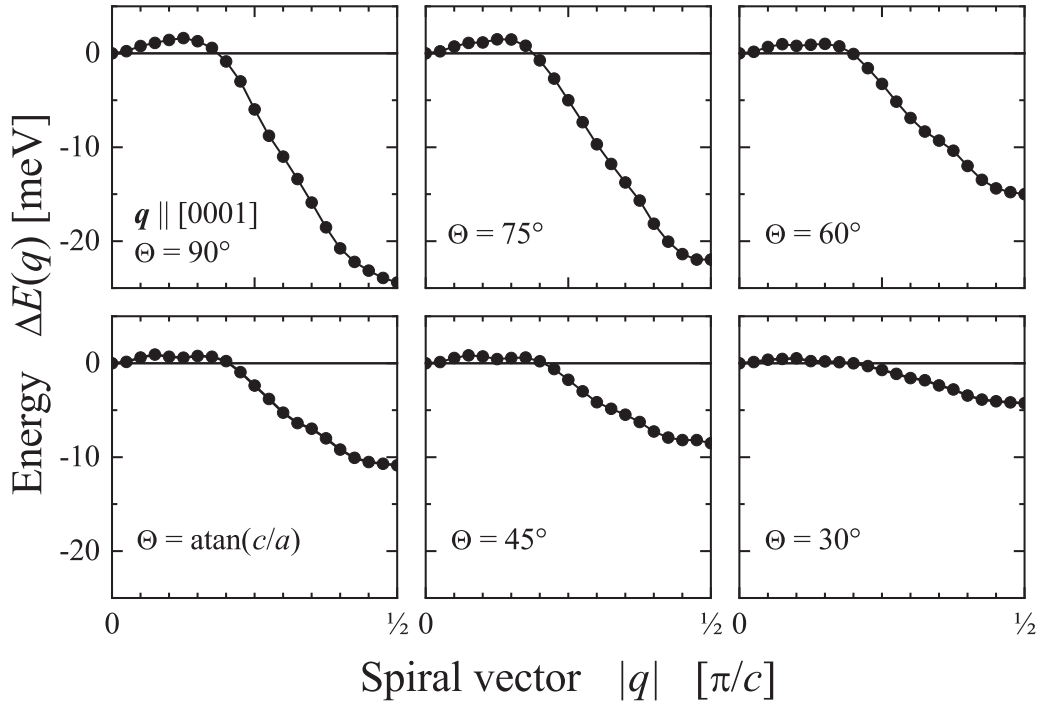


FIG. 9. Energies of antiferromagnetic cone spirals.

thin films [9]. The reason is that thin films generally have a shape (or dipolar) anisotropy which is distinct from the bulk material.

The electronic structure of MnPtGa in the canted magnetic state is illustrated in Fig. 12 by means of the density of states. Displayed are the atomic and the spin resolved contributions to the density of states ($n(E)$). The total spin magnetic moment amounts to $m_s = 6.22 \mu_B$, and the total orbital moment is $m_l = 0.08 \mu_B$. Both are parallel to the c axis.

It is found that the Ga atoms contribute mainly to the low-lying states at energies below -7 eV. Those states appear below a hybridization gap and are mainly of s character. It is also seen that the Pt d states are almost completely occupied and are located close to the bottom of the valence states with high densities in the energy range from -2 to -5 eV. The comparison of the site- and spin-resolved contributions reveals that the magnetic state is dominated by the localized Mn d states. These states exhibit an exchange splitting of approximately 3.4 eV, when estimated from the associated maxima of $n(E)$. The spin and orbital magnetic moments at the two Mn atoms are given by the vectors $\vec{m}_s^{\text{Mn}} = (\pm 2.19, 0.00, 3.12) \mu_B$ and $\vec{m}_l^{\text{Mn}} = (\pm 0.014, 0.00, 0.02) \mu_B$, respectively. The resulting overall, total (spin + orbital) magnetic moment at the Mn atoms is $m^{\text{Mn}} = 3.84 \mu_B$, which is only slightly higher than the values found for the collinear ferromagnetic and antiferromagnetic states. The net ferromagnetic moment, that is, the magnetic moment along c , reported in Ref. [5], ranges from 2.6 to 3.1 μ_B for temperatures between 10 and 200 K, which is close to the calculated value for the canted state.

More details of the electronic structure of the canted state are shown in Fig. 13 together with a comparison to the ferromagnetic state. The band structures are very similar, with some details shifted in energy or momentum space.

Differences in the obtained band crossings are due to the different symmetries of the two cases, that are the magnetic space group $P 6_3/mm'c'$ in the ferromagnetic and $P 2_1/m.1$ in the canted magnetic state. Such differences are easiest seen above the Fermi energy at the K point of the band structures. Various symmetry-related differences are also observed for the band positions and splitting at the Γ point. However, the exchange splitting—which is seen at the bottom of the bands at the M point—remains nearly unchanged (compare also Fig. 6, where this splitting is absent in the paramagnetic and antiferromagnetic states).

IV. SUMMARY

From the calculation of collinear magnetic order, it is found that the equilibrium energy of the antiferromagnetic order is slightly lower compared to the ferromagnetic order. This gives a hint that the low-temperature magnetic order state may be antiferromagnetic rather than ferromagnetic. The calculated Curie temperature is higher compared to the Néel temperature. Therefore, it can be concluded that the high- and low-temperature magnetic orders are different, and a magnetic phase transition may appear.

Calculations for noncollinear magnetic states reveal spiral or canted spin order. The spin spirals exhibit—independent of the initial spin order—the lowest energies for q along the [0001] direction. They give some hints on frustrated magnetism as the energies do not change much over wide q ranges. The ferromagnetic cone spiral has a minimum for a cone angle close to $\arctan(c/a)$. In both—ferromagnetic and antiferromagnetic—types of cone spirals, the energy minimum appears at $q = 0.5\pi/c$. However, various local minima are observed for the ferromagnetic spirals, pointing to metastable configurations. Therefore, a static canting of

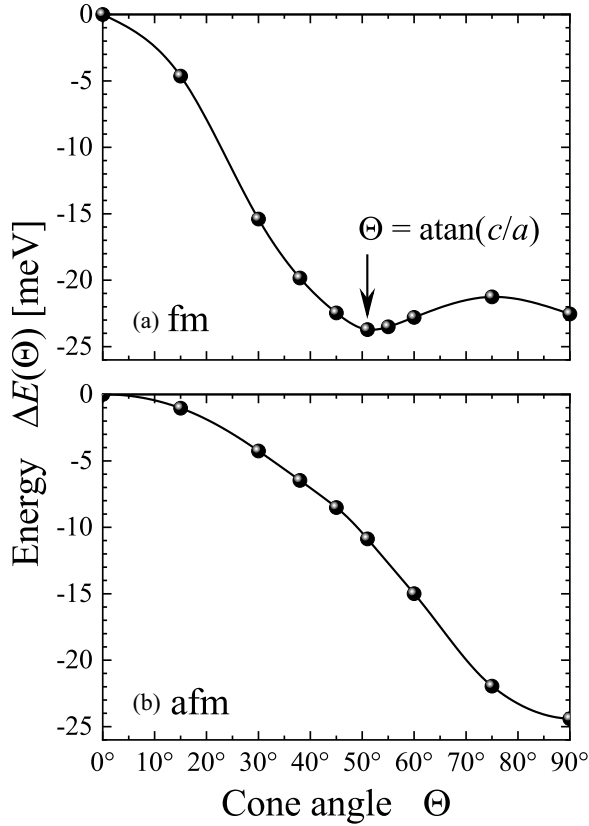


FIG. 10. Minimum energies of the cone spirals. Compared are the minimum spiral energies at $q = 0.5\pi/c$ for initial ferromagnetic (fm) and antiferromagnetic (afm) spin order.

the magnetic moments is possible. From the calculations for canted spin order, the lowest energy is found for a canting angle of about 38° . Thus, the ferromagnetic (parallel aligned) part of the magnetic moments is along the c axis, whereas the antiferromagnetic (antiparallel aligned) parts appear in successive Mn planes.

Overall, it is found that the energies for the various states—ferromagnetic, antiferromagnetic, spiral, canted—are all rather close together. This is in good agreement with the different types of magnetic order reported in the literature, in particular in Ref. [5]. Mostly, the energy differences are in the order of not more than 30 meV, independent of the used method. From this observation, it can be deduced that a particular magnetic state may be forced when an external field is applied. This includes more complicated noncollinear states as, for example, skyrmions or antiskyrmions. Considering the effect of temperature, a frustrated state frozen in a spin glass might be changed into a canted state already when a very small magnetic field is applied at finite temperature.

ACKNOWLEDGMENTS

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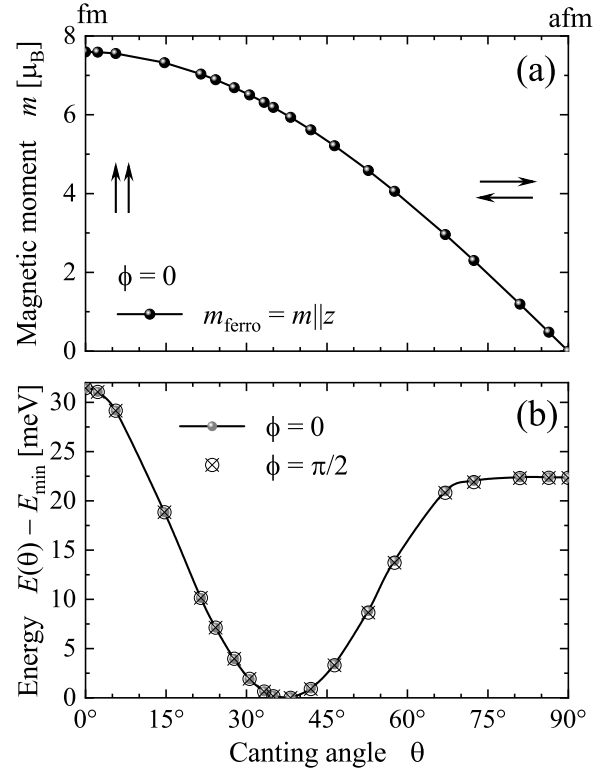


FIG. 11. Magnetic moments and energies in the canted, non-collinear magnetic state of MnPtGa. The angle θ was varied with respect to the [0001] direction at fixed $\phi = 0$ and $\phi = \pi/2$. The energy minimum appears at $\theta_{\min} = 37.82^\circ \pm 0.15^\circ$. Note that the alignment of the magnetic moments is along the hexagonal z axis for ferromagnetic type order, whereas it is in the basal plane for antiferromagnetic type order.

DATA AVAILABILITY

The data supporting this study's findings are available within the article.

APPENDIX A: MAGNETIC ANISOTROPY IN HEXAGONAL SYSTEMS

The general theory of the magnetocrystalline anisotropy is described in textbooks [52,53]. In most practical cases, only the uniaxial term of the magnetic anisotropy is given for hexagonal or tetragonal systems. However, this does not reflect the symmetry of the crystal structure. For hexagonal crystals, the uniaxial anisotropy constant may be calculated from the energy difference for magnetization along the c axis and an in-plane direction e.g., $K_u = E^{10\bar{1}0} - E^{0001}$, which is not unique because the in-plane Cartesian orthogonal coordinates x and y are not equivalent in hexagonal systems. In the following terms up to sixth order are investigated to account for the sixfold rotational symmetry of hexagonal systems. The extensions on higher order terms in the angular dependence of the magnetic anisotropy made here are similar to those for tetragonal systems reported in Ref. [54].

Care must be taken about the directions that have to be taken when calculating the magnetocrystalline energy from the first-principles energies using different computer

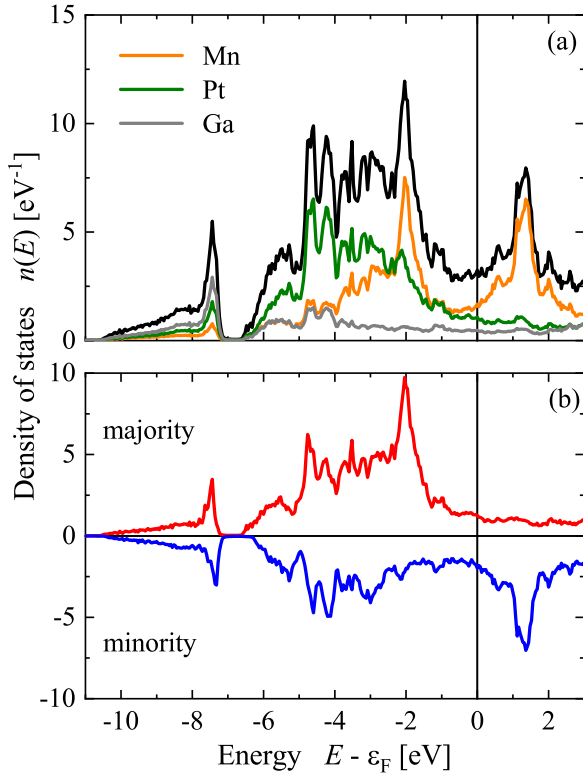


FIG. 12. Electronic structure of MnPtGa in the canted magnetic state with $\theta = 35^\circ$. (a) shows the site resolved and (b) the spin resolved density of states with respect to the primitive cell containing six atoms.

programs. For instance, the direction of magnetization (quantization axis) may be given either in terms of lattice vectors or Cartesian vectors, which differ in hexagonal systems. The three lattice vectors for the hexagonal structures are defined by: $\vec{g}_1 = (\sqrt{3}, -1, 0)a/2$, $\vec{g}_2 = (0, 1, 0)a$, and $\vec{g}_3 = (0, 0, 1)c$, that is, the vectors along x , y , and z are given by the combinations of lattice vectors $2\vec{g}_1 + \vec{g}_2$, $\vec{g}_2$, and $\vec{g}_3$, respectively. In other notation, using $hkil$ as indices, one has

$$\begin{aligned}\vec{g}_{10\bar{1}1} &= (\sqrt{3}, -1, 0)a/2, \\ \vec{g}_{01\bar{1}1} &= (0, 1, 0)a, \\ \vec{g}_{0001} &= (0, 0, 1)c,\end{aligned}\quad (\text{A1})$$

and the basis vectors in Cartesian coordinates are derived from

$$\begin{aligned}\vec{x} &= (2\vec{g}_{10\bar{1}1} + \vec{g}_{01\bar{1}1})/\sqrt{3}, \\ \vec{y} &= \vec{g}_{01\bar{1}1}, \\ \vec{z} &= \vec{g}_{0001}.\end{aligned}\quad (\text{A2})$$

Using a series expansion up to the sixth order in $\sin(\theta)$, the magnetocrystalline energy of a hexagonal system is expressed as

$$E_{\text{crys}}^{\text{hex}} = \sum_{\nu=0}^3 K_{\nu} \sin^{2\nu}(\Theta) + K_4 \sin^6(\Theta) \cos(6\Phi). \quad (\text{A3})$$

In the following, the lower index “crys” will be omitted, and the energies are indexed only by directions or by “hex.” From the high-symmetry directions $[h, k, i, l]$, one has for

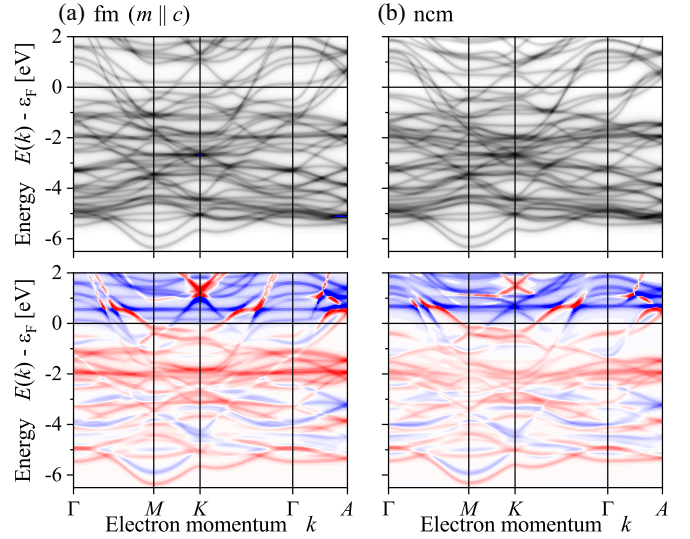


FIG. 13. Comparison of the band structures of MnPtGa in the ferromagnetic (fm) state with m along the c axis (a) and noncollinear, canted magnetic state (ncm) with $\theta = 35^\circ$ (b). Plotted are the total Bloch spectral function in gray and the spin-resolved Bloch spectral functions with majority electron states in red and minority states in blue.

variations of h or k (where $i = -(h + k)$) at fixed $l = 1$:

$$\begin{aligned}E^{0001} &= K_0, \\ E^{10\bar{1}1} &= \sum_{\nu=0,3} K_{\nu} \sin^{2\nu}(\Theta^{10\bar{1}1}) - K_4 \sin^6(\Theta^{10\bar{1}1}), \\ E^{20\bar{2}1} &= \sum_{\nu=0,3} K_{\nu} \sin^{2\nu}(\Theta^{20\bar{2}1}) - K_4 \sin^6(\Theta^{20\bar{2}1}), \\ E^{02\bar{2}1} &= \sum_{\nu=0,3} K_{\nu} \sin^{2\nu}(\Theta^{02\bar{2}1}) - K_4 \sin^6(\Theta^{02\bar{2}1}).\end{aligned}\quad (\text{A4})$$

Instead of $E^{20\bar{2}1}$ and $E^{02\bar{2}1}$, one can use $E^{01\bar{1}1}$ and $E^{21\bar{3}1}$, respectively:

$$\begin{aligned}E^{01\bar{1}1} &= \sum_{\nu=0,3} K_{\nu} \sin^{2\nu}(\Theta^{01\bar{1}1}) - K_4 \sin^6(\Theta^{01\bar{1}1}), \\ E^{21\bar{3}1} &= \sum_{\nu=0,3} K_{\nu} \sin^{2\nu}(\Theta^{21\bar{3}1}) + K_4 \sin^6(\Theta^{21\bar{3}1}).\end{aligned}\quad (\text{A5})$$

Using $z = c/a$, the various angles are found from $\Theta^{10\bar{1}1} = \Theta^{01\bar{1}1} = \arctan(1/z)$, $\Theta^{20\bar{2}1} = \Theta^{02\bar{2}1} = \arctan(2/z)$, and $\Theta^{21\bar{3}1} = \arctan(\sqrt{3}/(2z))$. Note that the direction $[21\bar{3}1]$ is along the x axis [see Eq. (A2)], whereas $[01\bar{1}1]$ is along y . Alternatively, $E^{10\bar{1}2}$ with $\Theta^{10\bar{1}2} = \arctan(1/(2z))$ may be used (and similar by increasing l at fixed h, k).

For in-plane vectors with a vanishing z component, one finds $\Theta = \pi/2$ such that $\sin^n(\Theta) = 1$ and thus for two perpendicular in-plane directions because $\cos(n\pi) = 1$ and

$\cos((2n-1)\pi) = -1$ ($n \in \mathbb{N}_0$):

$$\begin{aligned} E^{10\bar{1}0} &= \sum_{i=0,3} K_i - K_4, \\ E^{01\bar{1}0} &= \sum_{i=0,3} K_i - K_4, \\ E^{11\bar{2}0} &= \sum_{i=0,3} K_i - K_4, \end{aligned} \quad (\text{A6})$$

keeping in mind that $\phi_{10\bar{1}0} = -\pi/6$ and $\phi_{01\bar{1}0} = \pi/2$ or $\phi_{21\bar{3}0} = 0$ and $\phi_{12\bar{3}0} = \pi/6$. Further, it is found, for example, that $2K_4 = E^{12\bar{3}0} - E^{10\bar{1}0}$, or $E^{21\bar{3}0} - E^{01\bar{1}0}$, etc. Instead of $E^{11\bar{2}0}$, one can also use one of the following energy relations:

$$\begin{aligned} E^{1\bar{1}00} &= \sum_{i=0,3} K_i + K_4, \\ E^{12\bar{3}0} &= \sum_{i=0,3} K_i + K_4, \\ E^{21\bar{3}0} &= \sum_{i=0,3} K_i + K_4. \end{aligned} \quad (\text{A7})$$

Finally, the anisotropy constants K_i are found from the equations

$$K_0 = E^{0001}, \quad (\text{A8})$$

$$\begin{aligned} K_1 = -\frac{1}{12z^4} &[(60E^{0001} + 4E^{10\bar{1}1} - 64E^{10\bar{1}2})z^6 \\ &+ (36E^{0001} + 12E^{10\bar{1}1} - 48E^{10\bar{1}2})z^4 \\ &+ (12E^{10\bar{1}1} - 12E^{10\bar{1}2})z^2 \\ &+ 4E^{10\bar{1}1} - 4E^{10\bar{1}2} - 3E^{21\bar{3}0}], \end{aligned} \quad (\text{A9})$$

$$\begin{aligned} K_2 = \frac{1}{12z^4} &[(48E^{0001} + 16E^{10\bar{1}1} - 64E^{10\bar{1}2})z^8 \\ &+ (120E^{0001} + 56E^{10\bar{1}1} - 176E^{10\bar{1}2})z^6 \\ &+ (36E^{0001} + 72E^{10\bar{1}1} - 108E^{10\bar{1}2})z^4 \\ &+ (40E^{10\bar{1}1} - 25E^{10\bar{1}2} - 15E^{21\bar{3}0})z^2 \\ &+ 8E^{10\bar{1}1} - 2E^{10\bar{1}2} - 6E^{21\bar{3}0}], \end{aligned} \quad (\text{A10})$$

$$\begin{aligned} K_3 = -\frac{1}{12z^4} &[(48E^{0001} + 16E^{10\bar{1}1} - 64E^{10\bar{1}2})z^8 \\ &+ (60E^{0001} + 52E^{10\bar{1}1} - 112E^{10\bar{1}2})z^6 \\ &+ (12E^{0001} - 6E^{10\bar{1}0} - 6E^{21\bar{3}0} + 60E^{10\bar{1}1} \\ &- 60E^{10\bar{1}2})z^4 + (28E^{10\bar{1}1} - 13E^{10\bar{1}2} - 15E^{21\bar{3}0})z^2 \\ &+ 4E^{10\bar{1}1} - E^{10\bar{1}2} - 3E^{21\bar{3}0}] \end{aligned} \quad (\text{A11})$$

$$K_4 = -\frac{1}{2}[E^{10\bar{1}0} - E^{21\bar{3}0}], \quad (\text{A12})$$

where $z = c/a$. Further, equivalent energy terms from the above given equations may be used alternatively [for examples of equivalent energies, see Eqs. (A5) and (A7)].

The magnetocrystalline anisotropy energy (E_a) is the difference between the magnetocrystalline energy (here, E^{hex}) and the isotropic contribution, that is, the spherical part K_0 :

$$E_a = E^{\text{hex}} - K_0. \quad (\text{A13})$$

From this equation, the magnetocrystalline anisotropy energy may be positive or negative, depending on the direction and values of the K 's. This makes a visualization by plotting the three-dimensional distribution of $E_a(\vec{r}) = E_a(\theta, \phi)$ complicated. Therefore, the alternative anisotropy energy $E_{a'}$ with respect to the lowest energy is plotted most often, where

$$E_{a'} = E_a - \min(E_a) = E^{\text{hex}} - \min(E^{\text{hex}}), \quad (\text{A14})$$

which is still positive, although $E_a < 0$.

APPENDIX B: MAGNETIC ANISOTROPY AND APPLIED FIELD

The magnetic energy is altered if a magnetic field is applied. This has consequences for the determination of the magnetic anisotropy. The magnetic energy in an induction field $B_0 = \mu_0 H$ is [52,53,55]

$$E(\Theta, \Phi, \vec{H}) = E_{\text{ani}}(\Theta, \Phi) - M_s B_0 (\alpha_x \beta_x + \alpha_y \beta_y + \alpha_z \beta_z), \quad (\text{B1})$$

where $\alpha_i = \alpha_i(\Theta, \Phi)$ and $\beta_i = \beta_i(\theta, \phi)$ are the direction cosines of the magnetization and the applied field, respectively. Here, $E_{\text{ani}}(\Theta, \Phi)$ corresponds to the sixth-order anisotropy energy given by Eq. (A3). The spontaneous magnetization $M_s = M_s(T)$ depends on the temperature, as do the anisotropy constants K_i . The effect of the magnetic field on the magnetic energy distribution is illustrated in Fig. 14 for the case of ferromagnetic order (compare Fig. 4 fm).

APPENDIX C: DIPOLAR MAGNETIC ANISOTROPY

In non-cubic systems, the dipolar anisotropy does not vanish and also contributes to the magnetocrystalline anisotropy (see Refs. [54,56,57]). The complete Ewald lattice sum to find the dipolar energy reads

$$E_{\text{dip}} = \frac{\mu_0}{8\pi} \sum_i^{N_{\text{at}}} \sum_j^{N_{\text{at}}} \sum_{n=h,k,l}^{N_{h,k,l}} \left[\frac{\vec{m}_i \cdot \vec{m}_j}{|R_n|^3} - 3 \frac{(\vec{R}_n \cdot \vec{m}_i)(\vec{R}_n \cdot \vec{m}_j)}{|R_n|^5} \right], \quad (\text{C1})$$

where N_{at} is the number of atoms in the cell and $\vec{R}_n = \vec{r}_{ij} + \vec{R}_{hkl}$ and $\vec{r}_{ij} = \vec{r}_i - \vec{r}_j$ are the vectors between all atoms used in the triple lattice summation. Further, one has the important condition that $j \neq i$ for $\vec{R}_{hkl} = 0$. For hexagonal systems with lattice parameters a and c and the three lattice vectors $\vec{g}_i$ of Appendix A, one has $\vec{R}_{hkl} = h\vec{g}_1 + k\vec{g}_2 + l\vec{g}_3$, with $h, k, l \in \mathbb{Z}$.

The total number of lattice vectors is restricted by $\min(R_{\text{max}}, |R_{hkl}|)$ for a confining sphere with radius R_{max} . For a given sphere, the upper limits for the h, k, l may be estimated by $N_{h,k} > R_{\text{max}}/a$ and $N_l > R_{\text{max}}/c$. Alternatively, one may also restrict R_{hkl} to stay within a given cube. Wang and Holm [58] estimated the cutoff error when truncating the Ewald summation in Eq. (C1).

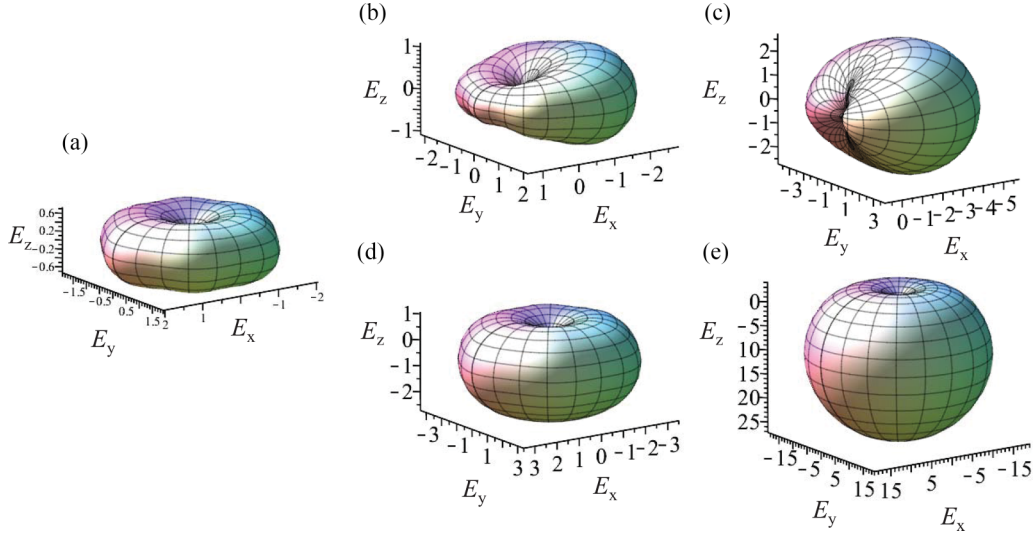


FIG. 14. Influence of an applied field on the magnetic energy distribution in the ferromagnetic case. Shown are the distributions for a field along x (b),(c) or z (d),(e) for low (b),(d) and high (c),(e) fields compared to a zero (a) field.

In general, the individual magnetic moments $\vec{m}_{i,j}$ do not necessarily have to be collinear. Their magnitude depends on the direction of magnetization $\hat{n} = \vec{M}/|M|$ such that not only the dipolar energy $E_{\text{dip}} = E_{\text{dip}}(\hat{n})$ but also the magnetic moments $\vec{m}_{i,j} = \vec{m}_{i,j}(\hat{n})$ become directionally dependent. To find the different magnitudes of the magnetic moments, one has to perform calculations that are fully relativistic or at least include the spin-orbit interaction as a perturbation.

Finally, the dipolar anisotropy is given by the difference between the energies for two different magnetization directions:

$$\Delta E_{\text{aniso}} = E(\hat{n}_2) - E(\hat{n}_1). \quad (\text{C2})$$

Two well-distinguished directions in hexagonal systems are $\vec{n}_1 = [0001]$ and $\vec{n}_2 = [01\bar{1}0]$, which are along the c axis and in the basal plane along a , respectively. Positive values indicate an easy dipolar direction that is along the $[0001]$ axis. It has a second-order angular dependence. Alternatively, a different in-plane direction, say $\vec{n} = [11\bar{2}0]$, $[1\bar{1}00]$, or $[10\bar{1}0]$, may also be used, and the results for inequivalent directions should be compared.

In a case that the magnetic moments are collinear and aligned along the magnetization direction, Eq. (C1) becomes

$$E_{\text{dip}}(\hat{n}) = \frac{\mu_0}{8\pi} \sum_i \sum_j m_i m_j \sum_n \left[\frac{\hat{n} \cdot \hat{n}}{|R_n|^3} - 3 \frac{(\vec{R}_n \cdot \hat{n})^2}{|R_n|^5} \right]. \quad (\text{C3})$$

The equation becomes further simplified because of $\hat{n} \cdot \hat{n} = 1$. The signs and values of the magnetic moments may still depend on the direction of magnetization. The lattice vectors $\vec{R}_n(i, j, h, k, l)$ still depend on the relative positions $(\vec{r}_i - \vec{r}_j)$ of the atoms in the basic cell. The last summation over n depends only on the structure and is called the magnetic dipolar Madelung sum M_{ij} :

$$M_{ij}(\hat{n}) = \sum_n \left[\frac{1}{|R_n|^3} - 3 \frac{(\vec{R}_n \cdot \hat{n})^2}{|R_n|^5} \right]. \quad (\text{C4})$$

The j -dependent part of Eq. (C3) is often introduced as the magnetic dipolar field $\vec{B}$. The dipolar field $\vec{B}_i$ at each atom of

the cell is calculated again from a direct Ewald lattice sum:

$$\begin{aligned} \vec{B}_i(\hat{n}) &= \frac{\mu_0}{4\pi} \sum_j \sum_n \frac{m_j}{|R_n|^3} [3(\hat{n} \cdot \hat{r}_n) \hat{r}_n - \hat{n}] \\ &= \frac{\mu_0}{4\pi} \sum_j m_j M_{ij}(\hat{n}), \end{aligned} \quad (\text{C5})$$

where $\hat{r}_n = \vec{R}_n/|R_n|$ represents the direction of the distance vectors. $\hat{r}_n$ depends on the position r_i of each atom in the primitive cell. Note, in some work, the double sum $\sum_i \sum_{j \geq i}$ is used instead of $1/2 \sum_i \sum_j$. The restriction on the sums is the same as for Eq. (C1).

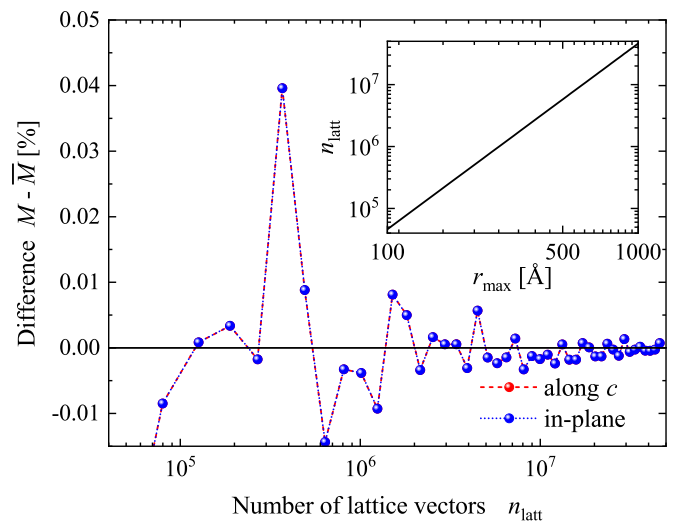


FIG. 15. Relative deviation of the Madelung sum as a function of the number of lattice vectors n_{latt} . Shown is the relative difference with increasing size of the confining sphere. The inset displays the dependence of n_{latt} on the radius of the confining sphere. ($\bar{M}$ is the mean value for $n_{\text{latt}} > 10^7$.)

Alternatively to (C1), the dipolar energy may be calculated from

$$E_{\text{dip}}(\hat{n}) = -\frac{1}{2} \sum_i \vec{m}_i \cdot \vec{B}_i. \quad (\text{C6})$$

Care must be taken that the Ewald lattice summations in the above equations converge. Figure 15 shows the behavior

of the magnetic dipolar Madelung sum $M_{ij}(\hat{n})$ of MnPtGa as a function of the radius R_{max} of the confining sphere. More than 40 million lattice vectors corresponding to $R_{\text{max}} \geq 1000 \text{ \AA}$ are needed to reach convergence. In the present work, $R_{\text{max}} = 1785 \text{ \AA}$ was used, corresponding to approximately 410 times the lattice parameter a . Note that the ferromagnetic dipolar Madelung sum is negative for $\hat{n}$ along the c axis.

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