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Dzyaloshinskii-Moriya interaction and magnetic anisotropies in Uranium compounds

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ABSTRACT

We report on the first-principles study of complex noncollinear magnetic structures in Uranium compounds. We contrast two cases. The first is the periodic magnetic structure of U_2Pd_2In with exactly orthogonal atomic moments, the second is an incommensurate plane spiral structure of UPtGe where the angle between atomic moments of nearest neighbors is also close to 90°. We demonstrate that the hierarchy of magnetic interactions leading to the formation of the magnetic structure is opposite in the two cases. In U_2Pd_2In, the magnetic anisotropy plays the leading role, followed by the Dzyaloshinskii-Moriya interaction (DMI) interaction specifying the chirality of the structure. Here, the interatomic exchange interaction does not play important role. In UPtGe the hierarchy of the interactions is opposite. The leading interaction is the interatomic exchange interaction responsible for the formation of the incommensurate spiral structure followed by the DMI responsible for the selected chirality of the helix. The magnetic anisotropy is very weak that is a prerequisite for keeping the distortion of the helical structure weak.

1. Introduction

The characteristic feature of Actinide compounds is complex interplay of physical interactions leading to a wide variation of physical properties [1]. This diversity is explained by the strongly varying extent of the localization of the 5f electrons. An important feature of the Actinides is strong spin-orbit coupling (SOC) that can lead to the formation of large atomic orbital moments and large magnetic anisotropy.

In the modern physics the effects caused by the SOC play very important role. In particular the Dzyaloshinskii-Moriya interaction (DMI) [2,3] responsible for the formation of the chiral magnetic structures, e.g., skyrmions [4,5], attracts much attention in the emerging field of spintronics. On the other hand, the role of the DMI in the physics of the U compounds has not been thoroughly investigated. In view of the strong SOC and wide range of magnetic properties the competition of different interactions is expected to play important role.

In this paper we contrast two Uranium compounds with complex noncollinear magnetic structures. By means of density-functional theory (DFT) calculations, we show that the hierarchy of the interactions in these compounds appears to be very different. One of the compounds is U_2Pd_2In with periodic magnetic structure formed by strictly orthogonal atomic moments [6]. The neutron scattering experiment has shown that the magnetically-compensated NC1 structure is the ground-state structure although three other noncollinear structures NC2-4 and collinear magnetic structures were also considered as candidates. The applied magnetic field revealed the presence of the first-order metamagnetic phase transition with substantial total magnetic moment.

The second compound is UPtGe where the magnetic structure is an incommensurate plane spiral structure, so-called cycloid, with almost orthogonal neighboring atomic moments [7].

2. U_2Pd_2In

The calculation of the energies of the NC1-4 states gives the following results. In agreement with experiment the lowest energy corresponds to the NC1 structure: $E_{NC2} - E_{NC1} = 1.48$ mRy/UC, $E_{NC3} - E_{NC1} = 5.94$ mRy/UC, $E_{NC4} - E_{NC1} = 7.77$ mRy/UC. Here UC is abbreviation for unit cell.

To discuss the energetics of the magnetic structures in terms of the competition of different types of interaction we map the system on the bilinear Hamiltonian of interacting atomic moments:

$$H = \sum_{IJ} \hat{S}_I A^{IJ} \hat{S}_J^T \tag{1}$$

where I and J number magnetic atoms, A^{IJ} are 3 by 3 matrices in the coordinate space, $\hat{S}_I$ is the unit vector in the direction of the Ith atomic moment. Matrix A^{IJ} can be written as the sum of the symmetric and

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antisymmetric parts [8]. The antisymmetric part can be recast in the form $\boldsymbol{D}_{IJ}\cdot[\hat{\boldsymbol{S}}_I \times \hat{\boldsymbol{S}}_J]$ and corresponds to the DMI. The symmetric part contains Heisenberg isotropic exchange and magneto crystalline anisotropy.

The questions we want to address are the following: (i) What is the physical reason for the formation of the magnetic structure with exactly orthogonal atomic moments? (ii) What makes the energy of the NC1 structure lower than the energies of other three structures with orthogonal atomic moments?

The symmetry analysis and the DFT calculations with neglected SOC [8] show that the exchange interactions cannot be responsible for the stability of the NC1 structure. Indeed, these calculations give for all four noncollinear structures NC1-4 exactly the same energy and the energy of the collinear ferromagnetic structure is lower by 8.23 mRy/UC than the energy of the noncollinear structures.

To estimate the DMI interaction we compare the energies of the pairs of the magnetic structures NC1 with NC2 and NC3 with NC4. Within these pairs, the atomic moments of each atom remain collinear that assumes that the local anisotropy energy of the atomic moment is unchanged. What does change is the 'local chirality' of the relative directions of the moments. The energetics of changing the chirality of the magnetic structure is governed by the DMI interaction. The obtained values of the energy differences are $E_{NC2} - E_{NC1} = 1.48$ mRy/UC and $E_{NC3} - E_{NC4} = 1.83$ mRy/UC. The fact that two differences are different reveals that the mapping of the electronic system on the simple bilinear Hamiltonian of atomic moments is not perfect. However, these results are sufficient to identify the place of the DMI in the hierarchy of the interactions in the system.

To estimate the strength of the local anisotropy of the atomic moments we consider the differences of the energy of the pairs of magnetic structures NC4,NC1 and NC3,NC2. Within these pairs the energies of both isotropic exchange interaction and DMI are the same and the energy differences must be assigned to the local anisotropy. We obtain the values of $E_{NC4} - E_{NC1} = 7.77$ mRy/UC and $E_{NC3} - E_{NC2} = 4.96$ mRy/UC. Again they are different in contrast to the expectation on the basis of the simple bilinear atomic model. However, the scale of the differences is distinctly larger than the scale of the DMI contribution given above.

On the basis of these calculations we can make the conclusion about the hierarchy of the interactions from the viewpoint of their contributions to the formation of the ground-state magnetic structure that can symbolically be expressed in the following form

exchange interaction < *DMI* < *magnetic anisotropy*

The local anisotropy selects NC1 and NC2 as candidates for the ground state while the DMI makes the final choice in favor of the NC1 structure. The isotropic exchange does not play significant role in the stabilization of the ground state magnetic structure (Fig. 1).

This physical picture is supported by the following calculation. We start with the ground state NC1 and rotate in the *xy* plane the atomic moment of the 1st atom until it becomes parallel to the moment of atom 3 (see Fig. 2). We calculate the band energy of the system as the function of the rotation angle. The curve has two close-in-energy minima at angles 0° and 180° and distinct maximum in the region of 90°. This curve shows again that the main energy scale is here the local magnetic anisotropy that makes energetically expensive to point the atomic moment in the direction orthogonal to the in-plane [11] axis.

The magnetic field applied along the [11] direction makes the

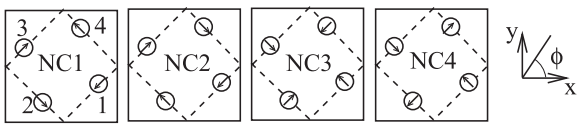

Fig. 1. Four noncollinear magnetic structures used in the analysis of the magnetic ground state of U_2Pd_2In.

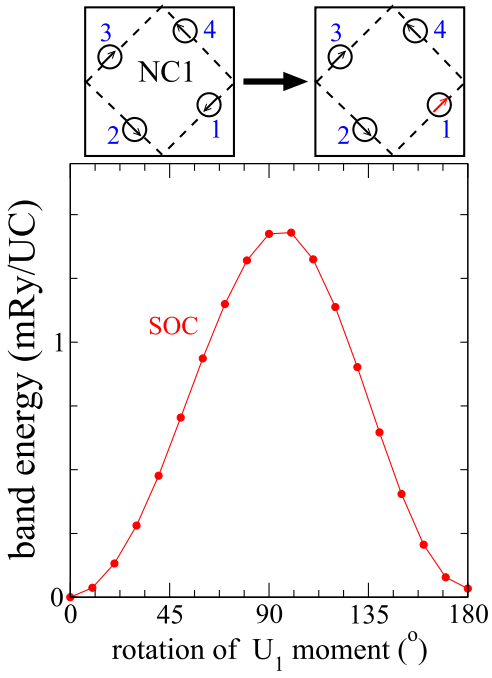

Fig. 2. The band energy of the magnetic structures obtained by the in-plane rotation of the direction of the magnetic moment of the U_1 atom starting with the NC1 structure. The two-minima curve is obtained in the relativistic calculations. In the upper part of the figure, the unit cells of the magnetic structures corresponding to the end points of the angular interval are depicted.

minimum at 180° deeper than the minimum at 0° that leads to the first-order phase transition from the NC1 state to the state with reversed moment of the 1st atom. This suggests an explanation of the nature of the metastable magnetic state and metamagnetic phase transition in the external magnetic field.

3. UPtGe

The magnetic structure of the UPtGe is an incommensurate spiral structure [7] (see Fig. 3). There are two important features we should remark about this magnetic structure [9]. First, it is unusual for the U compounds to have a spiral magnetic structure as a ground-state magnetic configuration. The reason for this is very strong magnetic anisotropy characteristic for these systems. The magnetic anisotropy makes regular rotation of the atomic moments characteristic for spiral structures energetically unfavorable since the moments have to assume various directions with respect to the crystallographic axes. The

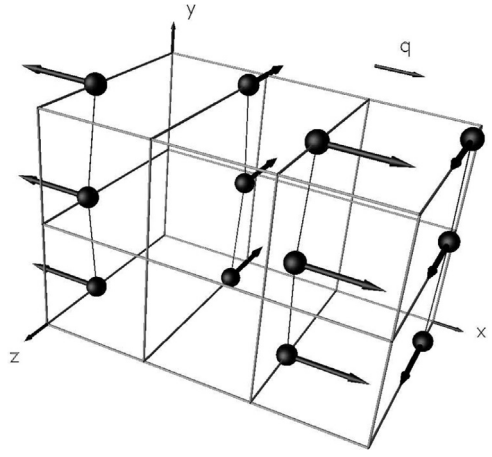

Fig. 3. Magnetic structure of UPtGe.

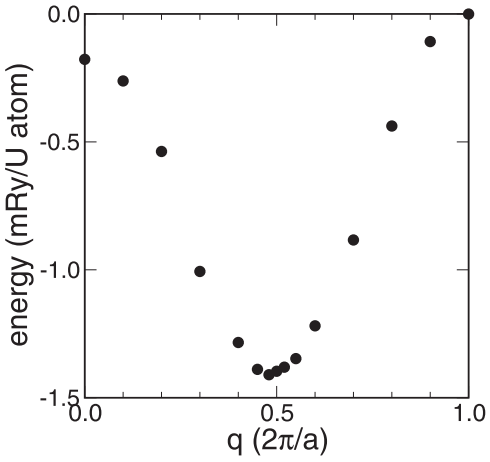

Fig. 4. Energy of UPtGe as a function of the wave vector of the spiral.

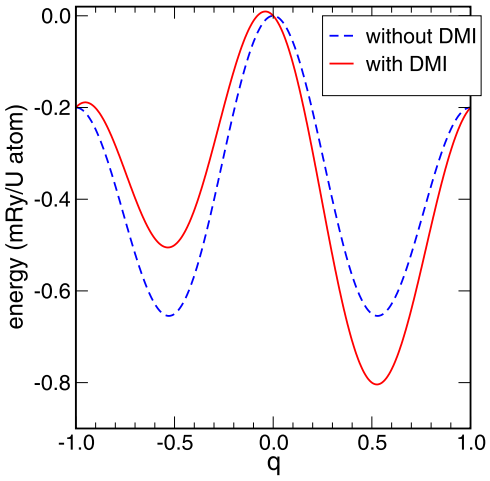

Fig. 5. Without DMI the energy as a function of **q** is symmetrical with respect to the reversal of the sign of **q**. The account for the DMI disturbs this symmetry.

magnetic anisotropy makes different directions inequivalent and the atomic moments tend to avoid the directions close to the hard axes in favor of the directions close to the easy axes. The second point is the experiment detection of the domains of one type of chirality whereas in a usual case the domains with opposite values of the spiral wave vector, **q** and **−q**, are equally present.

At the first stage of the calculations we switch off the SOC and calculate the energy of the system as a function of the spiral wave vector **q**. Because of the generalized translational symmetry of the nonrelativistic spirals such a calculation can be carried out easily and precisely for arbitrary value of **q** [10]. The result of the calculations shows that indeed the energy minimum is at an incommensurate **q** value (Fig. 4). Since in the calculation without the SOC the magnetic anisotropy and DMI are absent the origin of the spiral is the frustrated exchange interactions.

As remarked above one can expect that the strong magnetic anisotropy typical for the Uranium compounds strongly distorts the spiral structure. To understand why this apparently does not happen in the case of UPtGe we performed an estimation of the in-plane magnetic anisotropy. The estimation is performed by the comparison of the energies of the ferromagnetic configurations with atomic moments directed along perpendicular in-plane crystallographic axes. This estimation shows that in UPtGe the in-plane anisotropy is very small, less than 0.1 mRy/U atom. We emphasize that this calculation mast be performed with the SOC taken into account since the magnetic anisotropy is the consequence of the SOC. The small in-plane MAE despite strong SOC is a peculiar property of the UPtGe reflecting accidental compensation of the contributions of different electron states to the MAE.

To estimate the DMI energy we consider the supercell containing four U atoms and consider two commensurate spirals with pitch angles of 90° and −90° defining two opposite chiralities. Without the SOC the energies of these two structures are exactly equal to each other and the DMI vanishes. With the SOC taken into account two structures are inequivalent giving the energy difference of 0.3 mRy/U atom that provides an estimation of the DMI energy in the system. The DMI interaction disturbs the symmetry of the $E(q)$ function with respect to the reversal of the sing of q as shown in Fig. 5 and explains the observation of only **q** type of reflections and absence of the **−q** type of signals.

In summary, the analysis of the exchange interactions, DMI and MA allows to understand the physical origin of a rather unique among the U compounds spiral structure of UPtGe. In this case the hierarchy of the interactions takes the form

magnetic anisotropy < DMI < exchange interaction

and is opposite to the hierarchy of the same type of interactions in the noncollinear structure of U_2Pd_2In.

4. Conclusions

We report on the first-principles study of complex noncollinear magnetic structures in Uranium compounds. We contrast two cases. The first is the periodic magnetic structure of U_2Pd_2In with exactly orthogonal atomic moments, the second is an incommensurate plane spiral structure of UPtGe where the angle between atomic moments of nearest neighbors is also close to 90°. We demonstrate that the hierarchy of magnetic interactions leading to the formation of the magnetic structure is opposite in the two cases. In U_2Pd_2In, the magnetic anisotropy plays the leading role, followed by the DMI interaction specifying the chirality of the structure. Here, the interatomic exchange interaction does not play important role. In UPtGe the hierarchy of the interactions is opposite. The leading interaction is the interatomic exchange interaction responsible for the formation of the incommensurate spiral structure followed by the DMI responsible for the selected chirality of the helix. The magnetic anisotropy is very weak that is a prerequisite for keeping the distortion of the helical structure weak.

A natural question arises: Can a simple physical explanation be suggested for the drastic difference of the hierarchy of magnetic interactions in two Uranium compounds. Unfortunately, the answer is negative. The properties of effective magnetic interactions entering the spin Hamiltonian are complex cumulative consequences of the electronic structures of the materials where different parts of the electronic structure can contribute differently to the hierarchy relation. We emphasize, however, the important role of the crystal structure. For example, the DMI between two Uranium atoms can be active only if there is no inversion center transforming atoms into each other. This is relevant for both systems considered in the paper. On the other hand, the magnetic structures with exactly orthogonal atomic moments discussed in the case of U_2Pd_2In must be allowed by the symmetry of the lattice that simplifies the analysis of the magnetic interactions with the methods of first-principles calculations.

Acknowledgements

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