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Magnetic ordering in ZrNiAl-type crystal system

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Abstract

In this work, the theoretical studies, basing on the XY model, of possible magnetic ordering in ZrNiAl-type structure are presented. Our research are restricted to the ground state of XY Hamiltonian including interactions between nearest and next nearest magnetic moments. The influence of Dzialoshinsky–Moriya (DM) interaction is discussed as well. The results indicate several possible magnetic structures including simple ones of ferromagnetic or antiferromagnetic type. On the other hand, complicated magnetic structures described by commensurate as well as incommensurate propagation vectors were found as well. The resulting phase diagram of XY model is compared to experimentally determined magnetic structures of several compounds of ZrNiAl-type structure.

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1. Introduction

The RTX (R, rare earth; T, d-electron metal; X, p-electron element) rare earth intermetallic compounds exhibit a great variety of crystal and magnetic structures. In this work, we would like to pay attention to compounds, which crystallize in the hexagonal ZrNiAl-type structure (space group *P-62m*). Almost 30% of the RTX compounds crystallize in this crystal structure [1]. Neutron diffraction studies of the RTX compounds with ZrNiAl-type structure have been carried out for the last 15 years bringing interesting results [2–9]. To our best knowledge, studies within frame of XY model for ZrNiAl-type structure were not presented so far despite excellent experimental background.

In this structure, magnetic rare earth ions form a distorted kagomé lattice within the basal plane [7,10]. The coordination of magnetic moments leads to a geometrical frustration of magnetic interactions. The results of XY model for the triangle as well as the kagomé lattice are well known [11–13]; however, the lower symmetry of ZrNiAl lattice with respect to the kagomé lattice makes results derived for the latter one inapplicable.

2. Results and discussion

We have concentrated on the ground state of the following Hamiltonian:

$$H = -J_1 \sum_{i,i+1} \vec{S}_i \vec{S}_{i+1} - J_2 \sum_{i,i+2} \vec{S}_i \vec{S}_{i+2} - \vec{D} \sum_{i,i+1} \vec{S}_i \times \vec{S}_{i+1}$$

where J_1 and J_2 are exchange constants between nearest and next nearest spins, respectively, and D is a constant vector defining Dzialoshinsky–Moriya (DM) interaction [14]. The phase diagram calculations were made assuming all spins to be classical and taking into account spins lying within the basal plane only. For calculations including DM interaction the D vector was assumed to be perpendicular to the basal plane. The calculations were made for a honeycomb-like lattice containing 90 spins.

2.1. Nearest and next nearest interactions

When taking into account only J_1 and J_2 a quite complicated ground state phase diagram was obtained (see Fig. 1). Five phases can be established, however, it seems that only three of them are long-range ordered ones: ferromagnetic (F), antiferromagnetic of 120°-type (AF1) and antiferromagnetic modulated (AF2).

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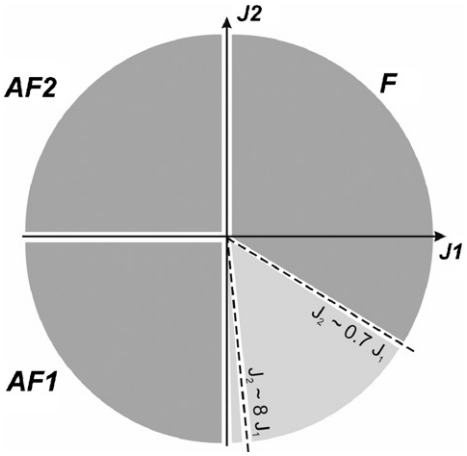

Fig. 1. Phase diagram of ZrNiAl-lattice with nearest (J_1) and next nearest (J_2) interactions. F, Ferromagnetic phase; AF1, antiferromagnetic phase of 120°-type and AF2, modulated antiferromagnetic phase. Long-range ordered phases are presented as dark areas.

A ferromagnetic phase is very common in RTX compounds of ZrNiAl-type structure, however, minority of them exhibits ferromagnetism with moments lying in the basal plane: NdPdIn [7], NdNiIn [8] and TbPdIn [15]. The phase exists when $J_1 > 0$ and $J_2 < 0.7J_1$. It is worth mentioning that only one exchange constant $J_1 > 0$ is required for establishing ferromagnetic phase.

The AF1 phase is presented in Fig. 2. As one can see magnetic moments form an angle of 120° between nearest and next nearest neighbors. The conditions J_1 and $J_2 < 0$ must be fulfilled in order to stabilize that phase. We made an observation that absence of negative J_2 leads to a possibility of arising other equienergetic (with respect to the AF1) states with no long-range order. The 120°-type phase is quite common in RTX compounds as a single phase or coexisting with other phases (e.g. ferromagnetic). Such ordering was found for: TbNiIn, DyNiIn, TbAuIn, DyAuIn, ErAuIn [8] and non-stoichiometric $HoNi_{0.8}In_{1.2}$ [9].

The second antiferromagnetic phase AF2 is shown in Fig. 2. In this case, a propagation vector (1/3; 1/3) describes the magnetic ordering within the basal plane. The magnetic moments still forms an angle of 120° between nearest neighbors, however, as one can see the second neighbors are ordered ferromagnetically. This structure is stable until $J_1 < 0$ and $J_2 > 0$. Similar structures with propagation vectors close or equal to (1/3; 1/3; 0) were observed for: DyAgSi, HoAgSi, TbAgGe [5], DyAgGe and HoAgGe [6].

When $J_1 > 0$ two phases with no long-range magnetic order were observed. One of them ranges from $-8J_1 < J_2 < -0.7J_1$ and is connected with destroying of the ferromagnetic order of nearest spins. When $J_2 < -8J_1$ the second neighbors are ordered antiferomagnetically forming an angle of 120° between each other.

2.2. Dzialoshinsky–Moriya interactions

Usually DM interaction is introduced when discussing the 3d magnetism. It was shown that in that case DM interactions originates from spin–orbit coupling [14]. DM interaction was considered as a small perturbation to the superexchange ($D/J \ll 1$).

On the other hand, DM interactions may be considered in case of indirect (RKKY) coupling between localized spins [16,17]. DM interaction was proposed to be oscillatory with manner of RKKY one, although the phase between DM and RKKY oscillations may differ [17]. Concluding, it is possible that in some cases D is even greater than J.

In Fig. 3, changes of magnetic ordering due to DM interaction are presented. The primary ferromagnetic structure is disturbed by introducing the D parameter. In all range of D/J_1 ratio a long-range ordered structures were observed (Fig. 3a–c). The structures were described by symmetric propagation vector k, which elements are shown in Fig. 3d. In case of $|D/J_1| = 0.1$, the propagation vector is given by $k = (0.02; 0.02)$ (see Fig. 3a). When $|D/J_1|$ ratio raises to 1, a magnetic structure described by $k = (1/6; 1/6)$ stabilizes (see Fig. 3b). When J_1 becomes negligible small with respect to D the propagation vector k approaching commensurate vector (1/3; 1/3) (see Fig. 3c). The asymptotically growth of k coordinates is shown in Fig. 3d.

Applying DM interaction to the AM1 or AM2 primary structure leads to departure from commensurate structures into "nearly" incommensurate. In this case, the structure is not

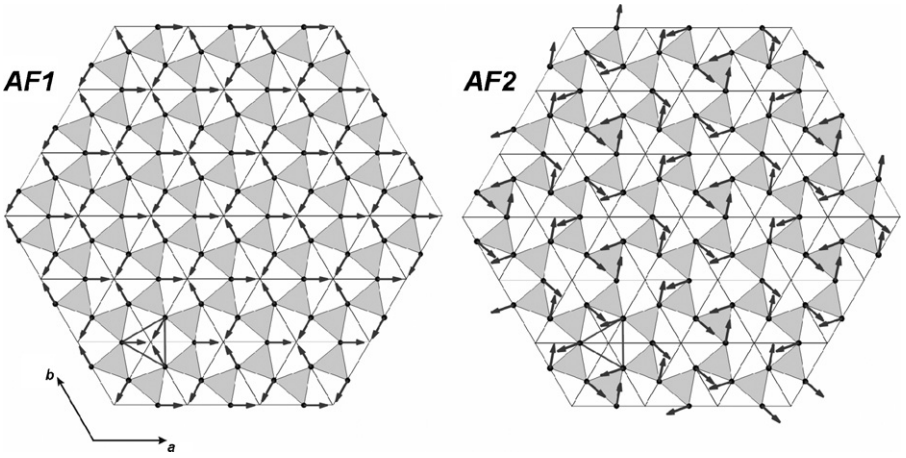

Fig. 2. Long-range ordered antiferromagnetic phases: AF1 is simple 120°-type phase and AF2 is phase with propagation vector (1/3; 1/3). The filled triangles represents the nearest spin coordination and one open triangle in the left bottom side of both clusters shows the next nearest spin coordination.

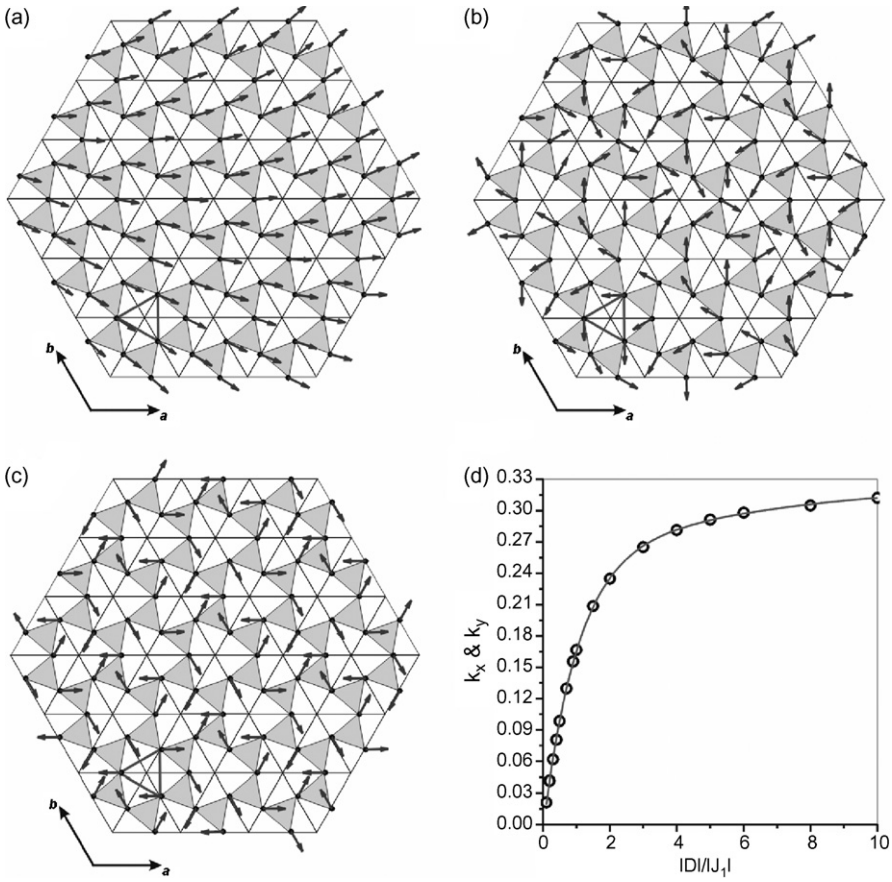

Fig. 3. Long-range order in case of presence of DM interactions while starting form ferromagnetic phase: (a) $J_1 = 1$ and $D = -0.1$; (b) $J_1 = 1$ and $D = -1$; (c) $D/J_1 \to \infty$; (d) propagation vector k vs. $|D|/|J_1|$ ratio. The filled triangles represents the nearest spin coordination and one open triangle in the left bottom side of both clusters shows the next nearest spin coordination.

exactly described by a given propagation vector, there are fluctuations of orientation's angle of spins up to 10° with respect to the ideal structure. This effect may be related to the limited size of the cluster.

3. Summary

The phase diagram of ZrNiAl-type lattice was investigated using XY model. Low symmetry of the lattice and frustration of magnetic interactions make variety of phases possible to exist. Taking into account only nearest and next nearest neighbors three long-range ordered phases are possible to develop. A very similar phases were observed in real compounds according to reported results of neutron diffraction experiments. Inclusion of the Dzialoshinsky–Moriya interaction rises an incommensurate behavior of magnetic structures. It seems that problem of magnetic ordering in ZrNiAl-type lattice is very complex. Consequently, further studies paying attention to influence of external magnetic field are required in order to compare theoretical predictions to single-crystal magnetization data.

Acknowledgements

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