Magnetic phase transitions in La$_{1-x}$Dy$_x$Mn$_2$Si$_2$ (0 ≤ x ≤ 1) compounds

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Abstract. Magnetic structures and properties of the quasi-ternary layered intermetallic La$_{1-x}$Dy$_x$Mn$_2$Si$_2$ (0 ≤ x ≤ 1) compounds were investigated using magnetic measurements on single crystals and neutron powder diffraction. It was shown that various magnetic structures and magnetic phase transitions, which are observed in the compounds with different Dy content, arise as result of changes in the intralayer Mn-Mn distances and competitions of the Mn-Mn, Mn-Dy and Dy-Dy exchange interactions.

1. Introduction

The ternary intermetallic $RM_2X_2$ compounds ($R$ is a rare earth, $M$ is 3d, 4d or 5d transition metal, $X$ is Si or Ge) were shown to exhibit very exciting physical properties ranging from superconductivity to heavy-fermion behavior [1]. The compounds are natural superlattices since they crystallize in the body-centered tetragonal ThCr$_2$Si$_2$-type structure (space group $I4/mmm$) consisting of alternating atomic layers of $\ldots R \ldots X \ldots M \ldots X \ldots R \ldots$ aligned perpendicular to the $c$-axis.

The $RM_2X_2$ compounds with $M$=Mn exhibit a unique set of magnetic phase transitions because of an unusual correlation between the intralayer Mn-Mn distance ($d_{\text{Mn-Mn}}$) and interlayer magnetic arrangement of the Mn magnetic moments [1, 2]. A critical distance for intralayer Mn atoms exists in the compounds and is considered to be equal to $d_c \approx 0.285$-0.287 nm at room temperature. As a rule, at $d_{\text{Mn-Mn}} > d_c$, the Mn layers are ordered ferromagnetically along the $c$-axis, whereas at $d_{\text{Mn-Mn}} < d_c$, they are ordered antiferromagnetically. The intralayer Mn-Mn distance can be changed by both external pressure and substitution of $R$ metal in the quasi-ternary compounds [3]. Within the layer the Mn magnetic moments form a canted ferromagnetic structure. The nature of this correlation remains unclear up to now.

In the $RM_2X_2$ compounds with magnetic rare earths, competition of the Mn-Mn, $R$-Mn and $R$-$R$ exchange interactions and anisotropy of the rare earth sublattice can strongly affect magnetic structures at low temperatures. For example, the of various exchange interactions competition results in the appearance of new types of magnetic structures and magnetic phase transitions in the ternary GdMn$_2$Ge$_2$ and DyMn$_2$Ge$_2$ compounds [4, 5, 6].

The aim of the present work was to study magnetic structure and magnetic properties of the La$_{1-x}$Dy$_x$Mn$_2$Si$_2$ (0 ≤ x ≤ 1) compounds in which there is a possibility to gradually change the intralayer Mn-Mn distances from $d_{\text{Mn-Mn}} > d_c$ (LaMn$_2$Si$_2$) to $d_{\text{Mn-Mn}} < d_c$ (DyMn$_2$Si$_2$) and the Dy-Mn and Dy-Dy exchange interactions by increasing Dy content.
2. Experimental details

The La$_{1-x}$Dy$_x$Mn$_2$Si$_2$ (x = 0, 0.1, 0.2, 0.27, 0.4, 1) compounds were obtained from initial components by induction melting in an argon atmosphere followed by annealing at T = 1293 K for one week. X-ray diffraction showed that all the compounds are single-phase and have the ThCr$_2$Si$_2$-type structure.

Quasi-single-crystalline samples in the form of plates were split from massive ingots. X-ray Laue analysis showed that the tetragonal c-axis for all samples was directed perpendicular to the plane of the plates, while in the plane of the plates a small disorientation of the a-axes was observed.

Magnetic measurements were performed using a SQUID magnetometer. Neutron powder diffraction measurements were carried out at the temperatures T = 4.2 K and 300 K using a D3 diffractometer with wavelength $\lambda = 0.24319$ nm at one of the horizontal channels of an IVV-2M reactor (the city of Zarechny). The calculation of neutron powder patterns was performed using the FULLPROF program.
3. Results and discussion

The replacement of La by Dy, which has a smaller atomic radius, leads to a gradual decrease in the lattice parameters and the unit-cell volume of the compounds. The intralayer Mn-Mn distance becomes close to the critical in the compound with \( x \approx 0.27 \).

Figures 1, 2 show the temperature dependences of magnetization of the compounds. For the compounds with the Dy content \( x < 0.2 \), the temperature dependences of the magnetization \( M(T) \) have the form typical of ferromagnets with a kink point at Curie temperature \( T_C \approx 300 \) K. For compounds with \( x > 0.2 \), an increase in the magnetization is observed at low temperatures, which evidences ordering of the Dy magnetic sublattice at temperatures below 50 K. For the compounds with \( x > 0.2 \), the \( M(T) \) dependences at temperatures \( T > 50 \) K have the form typical of antiferromagnets with maxima in the vicinity of the Neel temperatures \( T_N \approx 290 \) K.

The changes in the neutron diffraction pattern with increasing Dy concentration reflect the changes in the magnetic structure of the compounds at \( T = 4.2 \) K (Figure 3). Figure 4 presents the magnetic structures realized in the compounds at \( T = 4.2 \) K, which were determined using neutron diffraction experiments. In accordance with the \( M(T) \) dependences, four various magnetic structures exist in the compounds with different Dy content at \( T = 4.2 \) K. These structures can be described corresponding to the types of interlayer Mn-Mn magnetic ordering as F, F’, AF and F”. For LaMn2Si2, the ferromagnetic (F) interlayer Mn-Mn ordering exists. For the F’ structure, the ferromagnetic interlayer Mn-Mn ordering is retained and the ferromagnetic ordering in the Dy magnetic sublattice is observed for compounds with \( x = 0.1, 0.2 \). The Dy magnetic moments are antiferromagnetically coupled with Mn magnetic moments. In the AF structure, the Mn magnetic moments in the adjacent layers are ordered antiferromagnetically. For the F” structure, the Mn magnetic moments are ordered ferromagnetically both inside the layers and between adjacent layers. According to the \( M(T) \) dependences, the type of Mn-Mn interlayer ordering should change from ferromagnetic to antiferromagnetic at \( T > 50 \) K where the Dy magnetic sublattice is disordered.

In Figure 5, the data of the magnetic and neutron diffraction experiments are summarized in the \( x-T \) magnetic phase diagram. At temperatures \( T > 40 \) K, the Dy magnetic moments are disordered and the \( x-T \) diagram is similar to that for the La1-xSmxMn2Si2 quasi ternary compounds where Mn-Mn distance can be gradually changed by changing the Sm concentration [3]. The difference between the AF, F and AF’, F’ magnetic structures consists in the formation of order in the Dy magnetic sublattice for the AF’, F’ structures. The high temperature AF” magnetic structure corresponds to the antiferromagnetic in-plane ordering of the Mn magnetic moments [7].
With increasing Dy content up to \( x = 0.27 \), the intralayer Mn-Mn distance becomes close to the critical \( d_c \), which leads to the F'-AF', F-AF transitions. The AF'-F'' transition can be connected with the enhancement of negative Dy-Mn exchange interactions upon increasing Dy content. Thus, the changes in the type of the interlayer Mn-Mn ordering at \( T = 4.2 \) K in La\(_{1-x}\)Dy\(_x\)Mn\(_2\)Si\(_2\) are caused both by the existence of the critical intralayer Mn-Mn distance and competition of the interlayer Mn-Mn and Dy-Mn exchange interactions.

According to the performed magnetic measurements on the quasi single crystalline samples, all compounds show strong magnetic anisotropy with a \( c \)-axis easy direction. The magnetic anisotropy strongly increases with increasing Dy content as result of growth of contribution of the Dy magnetic anisotropy to the magnetic anisotropy of the compounds. For the compounds with \( x = 0.2 \) and \( x = 1.0 \), the irreversible magnetization processes are observed in magnetic fields below 10 kOe which can be connected with the first order field-induced transitions in the Dy and Mn magnetic sublattices.

In summary, the obtained results can be summarized as follows. The type of the Mn-Mn interlayer magnetic ordering at low temperatures changes from ferromagnetic to antiferromagnetic and becomes again ferromagnetic with increasing Dy content. The various magnetic structures and magnetic phase transitions, which are observed in the compounds with different Dy content, arise as result of changes in the intralayer Mn-Mn distances and competitions of the Mn-Mn, Mn-Dy and Dy-Dy exchange interactions.

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