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Critical scattering in the helimagnets $Fe_{1-x}Co_xSi$

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ABSTRACT

The critical scattering in the cubic noncentrosymmetrical helimagnets with Dzyaloshinskii–Moriya interaction $Fe_{1-x}Co_xSi$ ($x = 0.15$, 0.2, and 0.5) has been studied. The samples with small Co concentration ($x \leq 0.2$) are found to give the critical scattering with the q-dependence of the typical Lorentzian shape. The critical index of the inverse correlation length $\nu = 0.48 \pm 0.05$ is found for the compound with $x = 0.15$. The critical scattering above T_C has not been observed for $x = 0.5$. The critical index of the order parameter for the compounds with $x = 0.15$ and 0.20 are equal to $\beta = 0.220 \pm 0.005$ and 0.230 ± 0.007, respectively. The value of β could not been established for $x = 0.50$ because of the first order character of the phase transition. In our opinion, the change of the character of the phase transition from the weakly first order to the real first order type with increase of the Co concentration is related to the decrease of the anisotropy and increase of the itinerancy of the compounds under study.

1. Introduction

The cubic B20-type (space group $P2_13$) mixed compounds $Fe_{1-x}Co_xSi$ with $x \in [0.05; 0.8]$, the cubic noncentrosymmetrical helimagnets, order below T_C in a spin helix structure with a small propagation vector $k < 0.25\,nm^{-1}$ [1–4]. The features of their magnetic structure were recently studied and interpreted within the Bak–Jensen hierarchical model that takes into account the following interactions [5–7]: the exchange interaction, the isotropic Dzyaloshinskii–Moriya (DM) interaction, and the weak anisotropic exchange (AE) interaction. In full analogy to the magnetic structure of MnSi [8,9] and FeGe [10], the helicity is widely recognized to be induced by an antisymmetric DM interaction caused by the lack of a center of symmetry in the arrangement of magnetic atoms Fe and Co [5,11,12]. The weak AE interaction along with the cubic anisotropy mean for fixing the direction of the magnetic spiral. In spite of the numerous and detailed studies of $Fe_{1-x}Co_xSi$ compounds the critical scattering has not been a subject of investigations up to now.

There are, probably, two reasons for the lack of researcher's attention. Firstly, the low value of the ordered spin in these compounds [13,14] makes it difficult to collect an acceptable statistics in the critical scattering above T_C. Secondly, the low values of the wave vector k of these compounds require the SANS setup to be adjusted for the high resolution regime of measurements that, again, results in poor statistics of the detectable scattering intensity. In this paper we fill the gap in our knowledge and present the investigation of the critical phenomena for the samples $Fe_{1-x}Co_xSi$ with $x = 0.15$, 0.2, 0.5 by means of the polarized small angle neutron scattering.

The low temperature magnetic structure of these compounds was recently studied in Refs. [6,7]. The neutron diffraction experiments give the parameters of the magnetic system such as (i) the wave vector of the structure $\mathbf{k}$, (ii) the critical field H_{C1} marking the threshold of the anisotropy, and (iii) the critical field H_{C2} of the transformation from the spiral to the ferromagnetic phase. The results of these experiments were interpreted using the theory [15]. We evaluated the major interactions of the system: (i) the spin wave stiffness $A \approx g\mu_B H_{C2}/k^2$ characterizing the isotropic exchange interaction (Fig. 1(a)); (ii) the Dzyaloshinskii constant $SD \approx g\mu_B H_{C2}/k$ characterizing DM interaction (Fig. 1(b)) and (iii) the anisotropic energy $E_a \sim g\mu_B H_{C1}^2/H_{C2}$ (Fig. 1(c)). The monotonous dependence of A/a^2 on the concentration x demonstrates the absence of any correlation between this interaction and the critical temperature T_C shown in Fig. 1(c). The latter shows a slightly asymmetric bell-like shape as a function of x with a maximum at $x \sim 0.35$. On contrary, the x-dependence of the Dzyaloshinskii constant D (Fig. 1(b)) resembles quite well the behavior of T_C showing that the DM interaction is, likely, responsible for the critical temperature. The AE interaction was evaluated on the qualitative and quantitative levels. Qualitatively, it was shown that for $x < 0.2$ the helix wave vector $\mathbf{k}$ is clearly oriented along the $\langle 100 \rangle$ axes,

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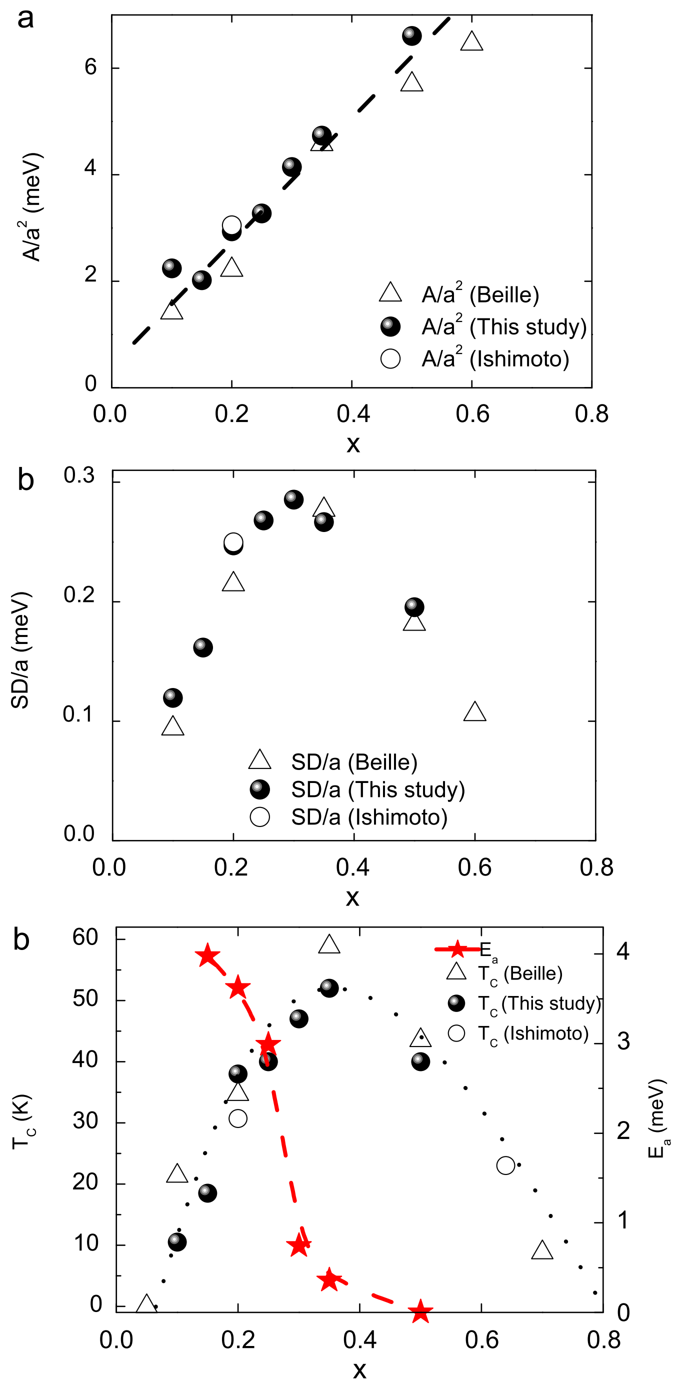

Fig. 1. Concentration dependencies of (a) the energy of the spin wave stiffness A/a^2, (△ [1,2], ○ [3,4], • is this study), (b) the Dzyaloshinskii constant SD/a, (c) the critical temperature T_C and the anisotropy energy E_a (★).

while for $x \geq 0.2$ it is almost randomly oriented. Since the anisotropy is responsible for the **k**-orientation then it is concluded to weaken with the increase of x. The quantitative estimations of the anisotropy are given in Fig. 1(c) (red stars). The AE interaction decreases sharply in the range of $x\sim0.25$. Being attributed to the electron spin–orbit scattering, it reveals the strength of bonds between the magnetically ordered electrons and the lattice. In our opinion, this break of the AE interaction is directly related to the electron transport properties of these compounds, which found to be half-metallic at $x<0.2$ and metallic at $x>0.2$ [1,16]. In this paper we study the influence of these factors on the critical phenomena in the $Fe_{1-x}Co_xSi$ compounds.

2. Experimental setup and samples

The SANS experiments were carried out at the SANS-2 scattering facility of the FRG-1 research reactor in Geesthacht (Germany). A polarized neutron beam with an initial polarization $P_0 = 0.93$, a neutron wavelength $\lambda = 0.58$ nm, a bandwidth $\Delta\lambda/\lambda = 0.1$, was used. The scattered neutrons were detected by a position sensitive detector with 256×256 pixels. A weak magnetic field (1 mT) guiding the polarization was applied horizontally in the sample's area. The scattering intensity was measured in the temperature range $8 \leq T \leq 60$ K with an accuracy better than 0.05 K.

A series of $Fe_{1-x}Co_xSi$ single crystals with $x = 0.15$, 0.2, 0.5 (at%) were chosen for the study. The disks with a diameter of 8 mm and a thickness of 1 mm were cut from large single crystals which were grown by the tri-arc Czochralski method. The sample–detector distances of 11 m (for $x = 0.15$), 9 m (for $x = 0.2$), 17 m (for $x = 0.5$) were used with appropriate collimation to cover scattering vectors q from 0.015 to 0.2 nm^{-1}. The critical temperature T_C for each sample has been obtained from the neutron scattering measurements as the temperature, where the magnetic Bragg reflection disappears. The obtained values are plotted in Fig. 1(c) along with those given in the other studies [1–4].

3. Critical scattering

The maps of the SANS intensities for the sample $Fe_{0.85}Co_{0.15}Si$ below and above $T_C = 18.65$ K are shown in Fig. 2 for the initial polarization P_0 along (a, c) and opposite (b, d) to the guiding field, respectively. Four Bragg peaks with $k = 0.172$ nm^{-1} and different intensities are visible at $T = 10$ K in Fig. 2(a, b). To observe four Bragg peaks at once is possible due to the large magnetic mosaic,

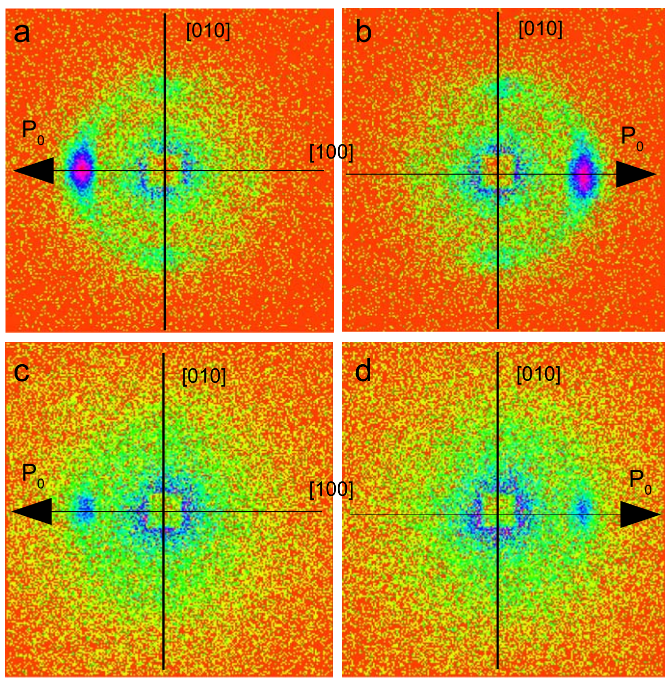

Fig. 2. The SANS maps of the sample $Fe_{0.85}Co_{0.15}Si$ at (a, b) $T = 10$ and (c, d) $T = 18.8$ K with the incident polarization P_0 along (a, c) and opposite (b, d) to the guiding magnetic field.

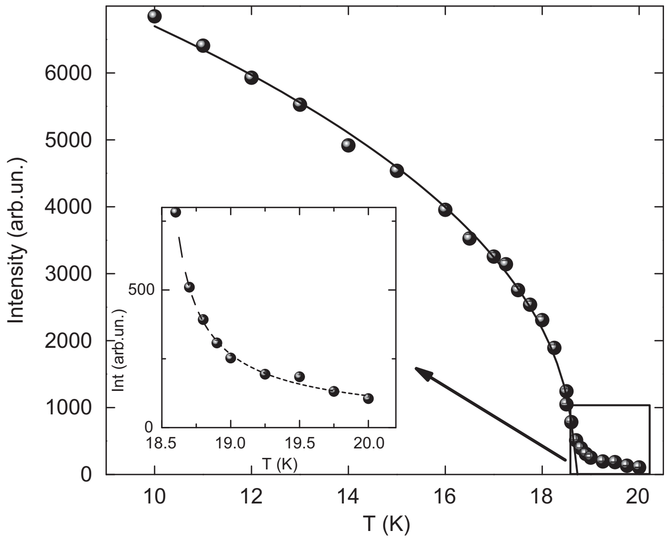

Fig. 3. Temperature dependence of the intensity of the [1 0 0] Bragg peak for the sample $Fe_{0.85}Co_{0.15}Si$. The inset shows the region above T_C more detailed.

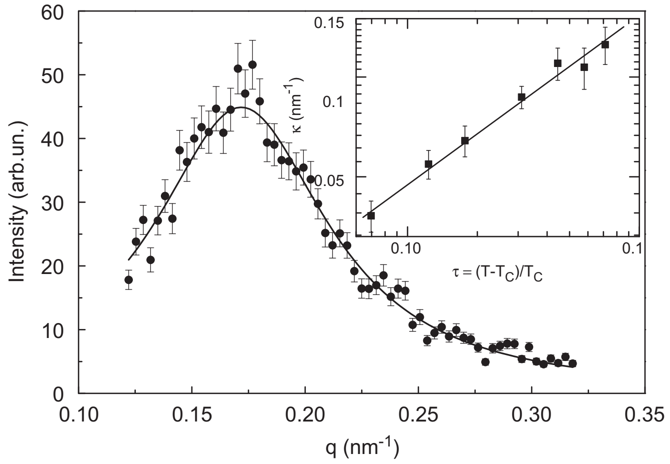

Fig. 4. Momentum transfer dependence of the scattering intensity for the sample $Fe_{0.85}Co_{0.15}Si$ at $T = 19$ K. Solid line is the fit to Eq. (1). Inset shows the inverse correlation length κ as a function of the reduced temperature τ.

as in ideal case the Bragg condition would be fulfilled for one reflection only. In our geometry it is the [1 0 0] peak. The well reserved Bragg peaks evidence that the AE interaction pin the wave vector $\mathbf{k}$ along the $\langle 1\,0\,0\rangle$ axes for the sample with $x = 0.15$. Interesting to note that the peaks at $\mathbf{k}\|[0\,1\,0]$ are polarization-independent, as $\mathbf{k} \perp \mathbf{P}_0\|[1\,0\,0]$. Reflections with $\mathbf{k}\|[1\,0\,0]$ depend on $\mathbf{P}_0$ as expected for helices with the Dzyaloshinskii vector $\mathbf{D}$ along $\mathbf{q}$. The intensity of the [1 0 0] peak is shown in Fig. 3 as a function of temperature. Its temperature variation obeys the scaling law $I \propto (-\tau)^{2\beta}$, where $\tau = (T - T_C)/T_C$ is the reduced temperature, $2\beta = 0.44 \pm 0.01$.

The scattering maps at $T = T_C + 0.15$ K in Fig. 2(c, d) still demonstrate weak spots along [1 0 0] axis, corresponding to the former Bragg peaks. They are well detectable within 0.3 K above T_C. Further increase of temperature leads to the appearance of the smeared ring of intensity with the maximum at $q = k$ attributed to the scattering on critical fluctuations. The scattering intensity at $q = k$ for $T > T_C$ is shown in the inset of Fig. 3. Its temperature dependence is described by the scaling law $I \propto \tau^{\gamma}$ with $\gamma = 0.79 \pm 0.04$.

The theoretical description of the critical neutron scattering for the cubic magnet with DM interaction was recently given in Ref. [17]. The neutron cross section in the mean field approximation for the paramagnetic phase ($T > T_C$) can be given in the following form:

$$\frac{d\sigma}{d\Omega} = r^2 \frac{T}{A[(q+k)^2 + \kappa^2]} \frac{k^2 + \kappa^2 + q^2 + 2\dfrac{D}{|D|}k\mathbf{q}\cdot\mathbf{P}_0}{(q-k)^2 + \kappa^2 + k^2 U(\hat{q}_x^4 + \hat{q}_y^4 + \hat{q}_z^4 - 1/3)}. \quad (1)$$

Here κ is the inverse correlation length of the spin fluctuations, $\mathbf{q}$ is the scattering vector, $\hat{q} = (\hat{q}_x, \hat{q}_y, \hat{q}_z) = \mathbf{q}/|q|$, A is the spin wave stiffness, $k = 2\pi/d$ is the helical wave vector (d is the spiral period), D is the Dzyaloshinskii constant, $\mathbf{P}_0$ is the incident polarization of neutrons, T is the temperature. Eq. (1) is well seen to have the Lorentzian form that gives a possibility to separate out the contribution of the critical fluctuations from the scattering on helix domain with the typically Gaussian shape. The theory was first applied to MnSi [17]. The results of polarized SANS experiments for MnSi and for $Fe_{0.85}Co_{0.15}Si$ look very similar and can be formulated as follows. (i) The diffuse scattering intensity above T_C looks strongly oriented along the incident neutron polarization. The DM interaction is responsible for this kind of scattering. (ii) The sum of the intensities for two opposite polarizations forms an anisotropic ring with weak spots. (iii) Below T_C these spots transform into the Bragg peaks originating from the helical structure.

The momentum transfer dependence of the scattering intensity for the sample $Fe_{0.85}Co_{0.15}Si$ at $T = T_C + 1.5 = 19$ K is presented in Fig. 4. The solid line shows the result of the fit of the experimental data to Eq. (1) with $k = 0.172\,\mathrm{nm}^{-1}$ and $\kappa = 0.064\,\mathrm{nm}^{-1}$. The whole set of the experimental data was fitted to Eq. (1). The inverse correlation length κ and the position of the maximum k were obtained as a result of the fit. The value of k does not change with temperature. The inverse correlation length κ is shown as a function of $\tau = (T - T_C)/T_C$ in the inset. The fit of the obtained dependence by the scaling law $\kappa \propto \tau^{\nu}$ (the solid line) gives a critical exponent of the inverse correlation length with $\nu = 0.48 \pm 0.05$. The resolution function with the width of $0.03\,\mathrm{nm}^{-1}$ was taken into account. The scaling relation between the indexes $\gamma + 2\beta = d\nu$ is satisfied within the error bars. It is worthwhile to note that the critical exponent of the order parameter $\beta = 0.22(1)$ is found for pure MnSi [17]. This suggests that this critical index is universal for the cubic magnets with the DM interaction.

The sample $Fe_{0.8}Co_{0.2}Si$ with $T_C = 38.2$ K has a behavior similar to that of the sample with $x = 0.15$. Thus, from the temperature variation of the Bragg intensity one can find that the critical exponent of the staggered magnetization is $2\beta = 0.46 \pm 0.07$ for the sample with $x = 0.2$. The critical scattering above T_C is though detectable but very weak and does not allow any reasonable treatment. At last, the sample $Fe_{0.5}Co_{0.5}Si$ does not show any critical scattering, the intensity disappears completely just above $T_C = 40.3$ K. The transition for this compound with $x = 0.5$ becomes clearly of the first order character. The change of the phase transition from the weakly first order to the first order type with increase of x is related to the decrease of the anisotropy (see Fig. 1(c)) which is correlated to the increase of the itinerancy of these compounds [14,16].

4. Concluding remarks

In this paper we have studied the critical behavior of the system $Fe_{1-x}Co_xSi$. The critical indices $\gamma = 0.79(4)$, $\nu = 0.48(5)$, and $\beta = 0.220(5)$ have been found for the $Fe_{0.85}Co_{0.15}Si$. The values of β for $x = 0.15$ and 0.2 are close to that of MnSi ($\beta = 0.22(1)$) [17]. This suggests that this critical index is universal for the cubic magnets with the DM interaction. The anisotropy in MnSi is found to be very strong of order of 15 meV [18] and the phase transition was identified as a weakly first order induced by critical fluctuations [19]. We suppose that the similar scenario of the phase transition can be implemented for the sample with $x = 0.15$ and for MnSi. But the character of the transition in $Fe_{1-x}Co_xSi$ is shown to change with the Co concentration x from the weakly first order type for $x < 0.2$ to the first order type for $x > 0.2$. The change in the critical behavior is correlated to the decrease of the anisotropy (Fig. 1(c)). The increasing of x leads to decrease of the anisotropy and to suppression of the critical fluctuations, thus, transforming the phase transition "more" into the first order type. This process takes place within the range $x \in [0.2-0.3]$, and in our view related to the half-metal to metal transition, which occurs in the same range of concentrations x [16].

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