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Magnetovolume effects and thermal expansion anomalies during a phase transition in a chiral ferromagnet MnSi with topological features of the electronic structure

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Within the framework of the theory of band magnetism and in the Heine model for the dependence of the electronic spectrum on volume, magnetovolume effects in the chiral quantum ferromagnet MnSi, with topological features of the electronic structure, are studied. New mechanisms of magnetovolume effects associated with the spin short-range order arising during a phase transition in fragments of right- and left-chiral spin helices with fixed Berry phases are considered. The calculated temperature dependences of the volumetric coefficient of thermal expansion (VTCE) describe the observed anomalies $\beta(T)$ and show that the disappearance of chiral spin charges associated with the Dzyaloshinsky–Moriya interaction is accompanied by a change in the sign of the VTCE. In this case, the fluctuation phase considered corresponds to that observed during small angle scattering of polarized neutrons.

Keywords: helicoidal ferromagnetism, chirality, spin fluctuations, electronic structure, thermal expansion.

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1. Introduction

The spin-orbit antisymmetric Dzyaloshinskii–Moriya (DM) interaction arises in strongly correlated band ferromagnet MnSi with a cubic non-centrosymmetric crystal structure without an inversion center (type B20). In neutron diffraction studies (see, e.g., [1,2]), twisting of magnetic moments $M_{\mathbf{v}}$ at nodes \mathbf{v} into so-called spin helices (Figure 1) with magnetic moments [3,4]

$$M_{\mathbf{v}}^{(x)} = M_S \cos(\mathbf{q}_0 \mathbf{v}), \quad M_{\mathbf{v}}^{(y)} = \pm M_S \sin(\mathbf{q}_0 \mathbf{v}), \quad (1)$$

and wave vector \mathbf{q}_0 directed along axis z , which is related to magnetic period $\lambda = \pi/|\mathbf{q}_0|$, is observed in the region of ferromagnetic long-range order. Jensen and Bak [5] have demonstrated that the formation of spin helices is caused by the competition between symmetric inhomogeneous exchange interaction and antisymmetric Dzyaloshinskii–Moriya interaction.

Ab initio calculations of the ground state for MnSi [6] yield a magnetic moment magnitude that differs notably from the one obtained in experiments. This discrepancy was rectified in [7], where it was attributed to anomalously large zero-point fluctuations in the ground state of MnSi. At the same time, a crossover of a magnetic first-order phase transition, which is accompanied by a change in the sign of the inter-mode interaction parameter in the Ginzburg–Landau functional, and a quantum transition leading to the suppression of zero-point spin fluctuations was found. Expressions for the root-mean-square amplitudes of zero-point and thermodynamic spin fluctuations,

which were determined using the fluctuation-dissipative theorem, were obtained in [7].

The results of neutron diffraction studies suggested that (1) is inapplicable to the fluctuation phase (Jensen–Bak „catastrophe“). It was demonstrated in [3,4] that vortex spin microstructures are topologically protected fragments of left-chiral spin helices [1,2]:

$$M_{\mathbf{v}}^{(x)} = M_S \cos(\mathbf{q}_0 \mathbf{v} + \phi), \quad M_{\mathbf{v}}^{(y)} = -M_S \sin(\mathbf{q}_0 \mathbf{v} + \phi), \quad (2)$$

and helices with chirality fluctuations [8]:

$$M_{\mathbf{v}}^{(x)} = M_S \cos(\mathbf{q}_0 \mathbf{v} + \phi), \quad M_{\mathbf{v}}^{(y)} = \pm M_S \sin(\mathbf{q}_0 \mathbf{v} + \phi). \quad (3)$$

Fragments of spin helices are found in the regions of spin correlations with fixed Berry phases [3,4]. Berry protectorates on the Fermi surface of MnSi were found in [9].

Magnetic contributions to the thermophysical properties at phase transitions in band magnets have been examined numerous times within the spin-fluctuation theory formulated in [10]. However, the thermodynamic model does not provide an unambiguous explanation for the observed complex pattern, since it has not been taken into account yet that the examined phase transition should be accompanied by volume effects. It was demonstrated in [11] that a second-order transition in MnSi should „break down“ to a first-order one due to the interaction of spin and lattice degrees of freedom. This is evidenced by smearing of the lambda-like anomaly of temperature dependences of the heat capacity and the coefficient of thermal expansion

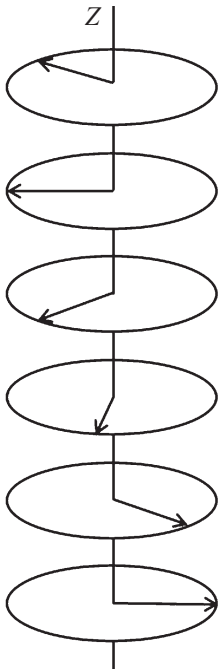


Figure 1. Flat helicoidal helix.

near the Curie temperature observed in experiments (see, e. g., [12]).

In the present study, the existing crossover model of quantum and thermodynamic first-order phase transitions in MnSi [3,4,7] is augmented by introducing the interdependence of local magnetization and volume, which induces magnetovolume effects. The obtained results are indicative of the presence of a hidden topological electronic transition (TEP) in the ground state of MnSi that proceeds due to the suppression of zero-point spin fluctuations in transition to the fluctuation phase. The experimentally observed anomalous magnetic contribution to the volumetric coefficient of thermal expansion (VCTE) is analyzed.

2. Model

Let us consider the electronic structure of the ground state of the MnSi band ferromagnet with account for the dependence of energy spectrum of strongly correlated d electrons $\varepsilon_{\mathbf{k}}$ on unit cell volume V , which is characterized by the well-known Heine formula [13]:

$$\varepsilon_{\mathbf{k}}(V) = \theta^{-1} \varepsilon_{\mathbf{k}}(V_0), \quad \theta(\equiv \theta(V)) = (V/V_0)^{5/3},$$

where $V_0 = 95.01 \text{ \AA}^3$ is the cell volume in the ground state.

Coulomb correlations in the system of d electrons are taken into account with the use of the Hubbard Hamiltonian with the added chiral DM interaction. Following [3,4] and using the coupling of spin (\mathbf{S}_{ν}) and charge (n_{ν}) density operators at node ν with the operators of secondary quantization of d electrons ($a_{\mathbf{k},\sigma}^+$, $a_{\mathbf{k},\sigma}$) in the state with a

quasi-momentum and spin, we present the Hamiltonian of the system in the form

$$H = H_0 - U \sum_{\nu} (S_{\nu}^2 - n_{\nu}^2/4) + \sum_{\nu, \nu'} d [\mathbf{S}_{\nu} \times \mathbf{S}_{\nu'}], \quad (4)$$

where

$$H_0 = \sum_{\mathbf{k}, \sigma} \varepsilon_{\mathbf{k}}(V) a_{\mathbf{k},\sigma}^+ a_{\mathbf{k},\sigma} \quad (5)$$

is the term of band motion of electrons, U is the Hubbard interaction constant, and d is the parameter of the DM interaction, which, owing to its relativistic smallness, is characterized by the mean field at node ν :

$$\mathbf{h}_{\nu}^{(D)} = \sum_{\nu'} [d_{\nu, \nu'} \langle \mathbf{S}_{\nu'} \rangle]. \quad (6)$$

The partition function of a system of strongly correlated electrons is characterized using the Matsubara technique for complex variables [14]:

$$Z = \text{Sp } T_{\tau} \left\{ - \int_0^{\tau^{-1}} d\tau H(\tau) \right\},$$

$$H(\tau) = \exp(H_0 \tau) H \exp(-H_0 \tau).$$

To examine the fluctuation pattern of the phase transition, we use the Hubbard–Stratonovich transformations [10] in calculation of the partition function:

$$\exp(-|A|^2) = \int d\xi \exp(-|\xi|^2 + A\xi),$$

where

$$(\xi = \xi, \eta, A = \mathbf{S}_{\nu}, n_{\nu}),$$

which allow one to reduce the many-body problem arising at the introduction of terms of Hamiltonian (4) quadratic in spin (\mathbf{S}_{ν}) and charge (n_{ν}) density operators to the analysis of electron motion in exchange and charge ξ and η -fields fluctuating in space and time

$$Z(\xi, \eta) = Z_0 \left\{ \int (d\xi d\eta) \exp(-\Phi(\xi, \eta)) \right\}, \quad (7)$$

where $Z_0 = Z(0, 0)$ is the partition function of band motion of d electrons with Hamiltonian H_0 ,

$$(d\eta d\xi) = d\xi_0 d\eta_0 \left[\prod_{q \neq 0, j} d\xi_q^{(j)} d\eta_q^{(j)} \right];$$

T is the temperature in energy units; $q = (\mathbf{q}, \omega_{2n})$, \mathbf{q} is the quasi-momentum; and ω_{2n} and ω_{2n+1} are the Matsubara Bose and Fermi frequencies.

It can be demonstrated that the free energy functional of electrons moving in fluctuating fields

$$\Phi(\xi, \eta) = \Phi_0(\xi, \eta) + \Delta\Phi, \quad (8)$$

consists, in accordance with [10], of the term characterized in the approximation of homogeneous local fields

$$\Phi_0(\xi, \eta) = \int_0^\beta d\tau \sum_{\nu \alpha (= \pm 1)} \int g_0(\varepsilon) \ln \left(1 + \exp T^{-1} \times (\mu - \varepsilon + \alpha c |\xi_\nu(\tau)| + ic \tilde{\eta}_\nu(\tau)) \right) d\varepsilon,$$

and corrections to the vertex parts of the second order of the series in ξ , which specify an anomalous dependence of the Stoner factor ($D_0(q, V, T)$) on the quasi-momentum and frequency in a magnetic phase transition:

$$\Delta\Phi = -T \ln \int_{-\infty}^{\infty} (d\xi, d\eta) \times \exp \left(- \sum_q (X_q |\eta_q|^2 + X_q |\xi_q - c^{-1} \mathbf{h}_q|^2) \right),$$

where $\mathbf{h}_q^{(\gamma)} = (\mathbf{h}_{\delta_{q,0}} + \mathbf{h}_q^{(D)}) \delta_{q,q}$, \mathbf{h} is a homogeneous external magnetic field,

$$D_0(q, V, T) = (1 - U \chi_q^{(0)}(q, V, T))^{-1},$$

$$\chi_q^{(0)}(q, V, T) = \sum \left(\frac{f(\varepsilon_k(V) - \mu) - f(\varepsilon_{k+q}(V) - \mu)}{(\varepsilon_k(V) - \varepsilon_{k+q}(V))} \right),$$

and μ is the chemical potential.

To obtain a consistent description of changes in the state of the spin system and volume, we take into account the well-known relation between the partition function and the free energy: $F = T \ln Z - \mu N$. The free energy of a system of strongly correlated electrons is supplemented by a term related to the elastic deformation energy of the crystal lattice with isothermal compressibility parameter K . Thus, the free energy is

$$F = T \ln Z(\xi, \eta) - \mu T + K(\Delta V)^2/2, \quad (9)$$

where $\Delta V = V - V_0$.

3. Magnetic state and magnetovolume effects

Let us consider the conditions of minimization of free energy (9) with account for the relation of the equilibrium values of spin and charge variables and the modulus of the wave vector of the spin structure with volume. We use here the saddle point conditions in fluctuating field variables

$$\xi_q^{(\gamma)} = r_q^{(\gamma)} \exp(\phi_{q,\gamma}),$$

$$r_q^{(\gamma)} = |\xi_q^{(\gamma)}| \text{ and } \xi_q^{(\gamma)} = |\xi_q^{(\gamma)}| \exp(\phi_{q,\gamma}), \quad (\mathbf{q} \neq \mathbf{q}_0),$$

tied to local magnetizations and pairwise spin correlators by known relations [14],

$$M_{\mathbf{q}}^{(\gamma)} = (U^{-1}) \xi_{\mathbf{q}}^{(\gamma)} - h_{\mathbf{q}}^{(\gamma)}, \quad |\xi_{\mathbf{q}}^{(\gamma)}|^2 = 2^{-1} (\langle T_\tau | S_{\mathbf{q}}^{(\gamma)}|^2 \rangle + 1),$$

γ is the index of spatial coordinate axes.

From the minima conditions, we derive an equation of state consisting of the equation for magnetization projections onto the coordinate system axes

$$M_{\mathbf{q}}^{(\gamma)} (D^{-1} + \kappa (M_S^2 + \langle m^2 \rangle / 3) + X(\mathbf{q}, 0)) = 2 \mathbf{h}_{\mathbf{q}}^{(\gamma)} / U, \quad (10a)$$

and the equation for volume

$$\omega = \Delta V(T) / V_0 = K^{-1} U^{-1} (\mathbf{M}_{\mathbf{q}_0}^2 + (2U)^{-1} \langle \delta M^2 \rangle + \langle m^2 \rangle). \quad (10b)$$

The resulting equations feature exchange enhancement factor

$$D = D(V, T) = (1 - U \theta \chi^{(\perp)} + 3^{-1} \theta \kappa \langle m^2 \rangle)^{-1}, \quad (10c)$$

inter-mode coupling parameter

$$\kappa = \kappa(V, T) = U \langle m^2 \rangle^{-2} (\chi^{(\perp)} - \chi^{(\parallel)}), \quad (10d)$$

mean square of local magnetization

$$M_S = \langle \delta M \rangle^{1/2}, \quad \langle \delta M^2 \rangle = \sum_q |\mathbf{M}_{\mathbf{q}}|^2,$$

and mean-square amplitude of thermodynamic zero-point and thermal spin fluctuations

$$\langle m^2 \rangle = \langle m^2 \rangle_0 + \langle m^2 \rangle_T = (4\pi U)^{-1} \times \sum_{q\gamma} \int_0^\infty (1/2 + f_B(\omega/T)) \text{Im} (D^{-1} + 2\kappa |M_{\mathbf{q}_0}^{(\gamma)}| + X_q)^{-1} d\omega, \quad (11)$$

where the term with the Bose–Einstein function corresponds to thermal fluctuations and the term with „1/2“ corresponds to zero-point ones.

Transverse and longitudinal susceptibilities found in (10d) are given by

$$\chi^{(\perp)} = \lim_{q \rightarrow 0} \sum \left(f(\varepsilon_k(V) - \mu) - f(\varepsilon_{k+q}(V) - \mu) / (\varepsilon_k(V) - \varepsilon_{k+q}(V)) \right),$$

$$\chi^{(\parallel)} = \lim_{q \rightarrow 0} \sum f(\varepsilon_k(V) - \mu) - f(\varepsilon_{k+q}(V) - \mu) / (\varepsilon_k(V) - \varepsilon_{k+q}(V)).$$

Solving equation of state (10a), we find that local magnetization and the magnetovolume effect (ΔV) arise in the considered system not only in the case of helicoidal

helices (1) with wave vector $|\mathbf{q}_0| = d/U^2 g(\theta, \mu)$ (long-range order region), but also in the fluctuation phase with fragments of spin helices (2), (3).

1. At negative values of the exchange enhancement factor (10c) and positive values of the inter-mode interaction parameter (10d), long-range order with chiral ferromagnetic helices is established in the studied system. At $T < T_c$ (Curie temperature) and $V < V_c (= V(T_c))$, this ordering leads to a relative volume change

$$\omega_1 = (V_c - V_0)/V_0 \approx K^{-1}U^{-1}[M_S^2 + \langle m^2 \rangle], \quad (12)$$

where local magnetization squared

$$\begin{aligned} M_S^2 &= M_S^2(V, T) \\ &= (2\kappa)^{-1} \left((D^{-1} + X(\mathbf{q}_0, 0))^2 - (d|\mathbf{q}_0|/U)^2 \right)^{1/2}. \end{aligned}$$

2. When the sign of the inter-mode interaction parameter changes, ferromagnetism loses its stability and short-range order is established with pairwise spin-spin correlators $\langle \mathbf{S}_\nu, \mathbf{S}_\mu \rangle \sim \exp(|\nu - \mu|/R_c)$. According to the phase transition theory, their radius is $R_c = (Ug(\mu, V)A)^{1/2}|D|^{-1/2}$, where parameter A characterizes the nonuniformity of the Lindhard function ($\chi_0(q) = \chi_0 + Aq^2$).

Fragments of spin helices (2), (3) with fixed Berry phases (ϕ) emerge within the spatial regions of spin correlations [3,4].

Note that one needs to distinguish between left-chiral ferromagnetic short-range order with $D < 0$ and the region of mixed (right and left) chirality of helices with $D > 0$ [3,4] found at $T_c < T < T_{extS}$ and volume ranging from V_c to $V_s (= V(T_s))$.

2a). In the region of chiral ferromagnetic short-range order, we obtain the solutions for left-chiral helices (2), which lead to a relative change in volume

$$\omega_2 = (V_s - V_c)/V_0 \approx K^{-1}U^{-1}[M_s^2 + \langle m^2 \rangle]. \quad (13)$$

2b). In the region of mixed chirality (3) arising from the D sign change at $T_s < T < T_{DM}$ within the interval from V_s to $V_{DM} (= V(T_{DM}))$, we find

$$\omega_3 = (V_{DM} - V_s)/V_0 \approx K^{-1}U^{-1}[M_s^2 + \langle m^2 \rangle_T]. \quad (14)$$

Here, the values of T_{DM} and V_{DM} correspond to the upper phase boundary of chiral spin short-range order.

As in [3], it may be demonstrated for the entire examined fluctuation phase that non-zero values of triple spin correlators, which are interpreted as chiral topological charges, arise in an external magnetic field with magnetization $M_0^{(z)}$:

$$\begin{aligned} \chi_c &= \sum_{\mathbf{v}_1, \mathbf{v}_2, \mathbf{v}_3} \langle \mathbf{S}_{\mathbf{v}_1} [\mathbf{S}_{\mathbf{v}_2} \times \mathbf{S}_{\mathbf{v}_3}] \rangle \\ &= \pm 2M_0^{(z)} M_S^2 (|\mathbf{q}_0| R_c)^{-1} \sin^2(|\mathbf{q}_0| R_c). \end{aligned}$$

Sign „–“ should be used in the region with fluctuations of left-chiral helices, and topological charges of both signs „+“ are found in the region of mixed chirality.

The emergence of topological charges implies the preservation of spin chirality effects in the fluctuation phase and provides direct evidence of TEP, which can only be associated with volume changes (magnetovolume effects) in the ground state of the spin short-range order phase.

In this case, chemical potential μ and the Fermi energy (equal to μ at $T = 0$) are determined by the conditions of electroneutrality for the numbers of s (p) and d electrons (written with account for the saddle point conditions in charge variable η_0):

$$N_e = N_S + \sum_{\alpha=\pm 1} \int d\varepsilon f(\varepsilon - \mu + \alpha U m) g(\varepsilon, V). \quad (15)$$

Here, N_e is the total number of electrons, N_S is the number of s electrons, $f(\varepsilon)$ is the Fermi–Dirac function, and $m = (\langle m \rangle_0^2 + M_s^2 + \langle m^2 \rangle_T)^{1/2}$.

4. Model of the electronic structure

In numerical analysis of the ground state with account for electroneutrality condition (15), we examine the density of electronic states found earlier in [4] using the GGA + U approximation in the Elk software package (<http://elk.sourceforge.net>).

The density of electronic states from [4] is shown in Figure 2 alongside with the Fermi energy shifts associated with volume changes (see Section 5 below). The results of numerical analysis with account for the DOS found in [4] reveal that, owing to a change in volume, the Fermi level in the ground state of the fluctuation phase falls into the Berry curvature region [9], and the inter-mode coupling parameter in this region is negative.

The boundaries of long-range order regions with helical spin helices and regions of the fluctuation phase

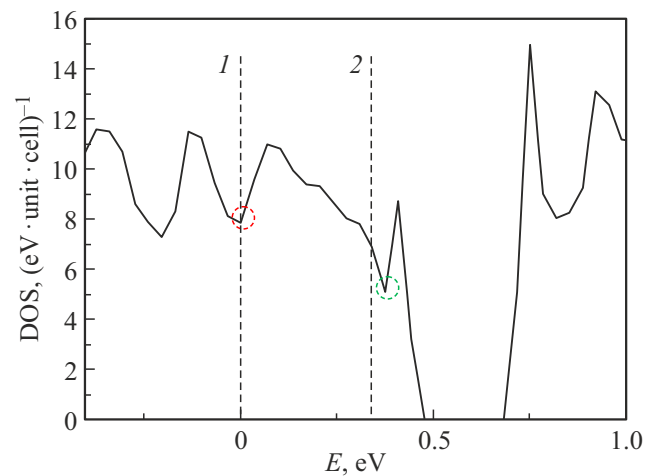


Figure 2. Fragment of the density of d electronic states in MnSi calculated in the GGA + U approximation [4]: 1 — position of the Fermi level in the ground state with zero-point spin fluctuations; 2 — position of the Fermi level in the ground state of the fluctuation phase.

are determined at a temperature shift of the chemical potential with account for electroneutrality condition (15). The paramagnetic phase, where local magnetization M_S^2 vanishes, is obtained by shifting the chemical potential level beyond the region with negative inter-mode coupling.

5. Magnetic contribution to thermal expansion

Comparing the obtained data on magnetic and volume effects with experimental results, we analyze the observed temperature dependences of VCTE, which provide not only supplementary, but also the most accurate techniques for experimental determination of temperatures and volumes.

The temperature dependence of the volumetric coefficient of thermal expansion was determined through the temperature derivative of the relative volume change (10b):

$$\beta = \partial \omega / \partial T.$$

Both magnetic (β_{mag}) and non-magnetic (phonon β_{oph} [15] and electronic β_{oeI}) contributions to VCTE were evaluated:

$$\beta = \partial \omega / \partial T = \beta_{\text{oeI}} + \beta_{\text{oph}} + \beta_{\text{mag}}. \quad (16)$$

Estimates of the phonon component ($\beta_{\text{oph}} \sim (T/T_D)^3$, where Debye temperature $T_D = 450$ K) and the contribution associated with electronic Fermi excitations ($\beta_{\text{oeI}} \sim (T/T_F)$, where Fermi temperature $T_F \sim 10^4$ K) demonstrate that they are negligible ($\beta(T) \approx \beta_{\text{mag}}(T)$) in the considered temperature range of the phase magnetic transition ($T < T_{\text{DM}} = 32$ K).

The magnetic contribution to VCTE was found numerically by solving the equations resulting from saddle point conditions. The effective mass approximation for the Lindhard function was used for calculating the amplitudes of spin fluctuations (11):

$$\begin{aligned} X(\mathbf{q}, \omega) &= U(\chi^{(0)}(0, 0) - \chi^{(0)}(\mathbf{q}, \omega)) \\ &= (A\mathbf{q}^2 - iB\omega\theta(\omega_0 - \omega)/|\mathbf{q}|), \end{aligned}$$

where Lindhard function parameters $A = 0.07$, $B = \pi/2.45$ were taken from [3,4]. These parameters were determined there by comparing the results of calculations of magnetic susceptibility to experimental data with experimental T_c values used as a parameter.

The numerically determined temperature dependence of the magnetic contribution to VCTE is shown in Figure 3. The results of calculation of the magnetic VCTE component reveal that the emergence of the fluctuation phase is accompanied by the suppression of zero-point spin fluctuations at Curie temperature (T_c) [7]. This leads to a β_{mag} anomaly near T_c ; a sharp $\beta_{\text{mag}}(T)$ increase associated with a change in volume (13) is observed in the fluctuation phase with fragments of helices (2) within an extremely narrow range from $T_c = 28.74$ to $T_S = 29.1$ K. A distinct „shoulder“

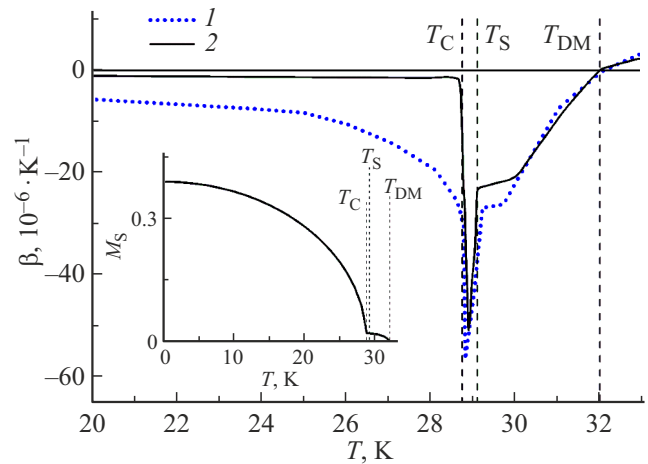


Figure 3. Temperature dependence of the volumetric coefficient of thermal expansion of MnSi: 1 — experimental data [12]; 2 — calculation of the magnetic contribution in the present study. The temperature dependence of local magnetization of MnSi is shown in the inset.

forms in the interval of weak temperature dependence of M_S^2 at $T_S < T < T_{\text{DM}}$ ($T_{\text{DM}} = 32$ K) in the region of mixed spin chirality ((3), (14)). According to equation of magnetic state (10a), an approximate quadratic dependence of local magnetization is established in transition to the paramagnetic phase:

$$M_S^2 \approx M_S^2(T_S)(1 - (T/T_{\text{DM}})^2),$$

and the negative VCTE becomes positive at T_{DM} .

Integrating the $\beta(T)$ dependence following from the considered model, we find that at long-range order phase temperatures $T < T_c$ $\omega_1 \approx 2.2 \cdot 10^{-5}$; $T_c < T < T_S$, $\omega_2 \approx 1.4 \cdot 10^{-5}$; and at $T_S < T < T_{\text{DM}}$ $\omega_3 \approx 3.4 \cdot 10^{-5}$.

6. Conclusion

Thus, the magnetization and volume changes in strongly correlated chiral ferromagnet MnSi with a crystal structure without an inversion center are interdependent. This results in chiral spin short-range order in the fluctuation phase, which is consistent with the results of experiments on small-angle scattering of polarized neutrons [1,2,8] and the observed VCTE anomalies.

Features of the VCTE temperature dependence arising in the fluctuation phase lead to the emergence of a lambda-like anomaly and a „shoulder“ in the dependence of negative $\beta_{\text{mag}}(T)$ and to a change in its sign when the chemical potential shifts beyond the region with negative inter-mode coupling.

It is obvious that the observed lambda-like anomaly and „shoulder“ in the temperature dependence of heat capacity at constant pressure [11], which differs from the heat capacity at constant volume by a correction proportional to β , have similar causes.

The phase transitions examined here using the example of a prototype spintronic topological material (MnSi) are also possible in doped Weyl semimetals (based, e.g., on CoSi [16]), where skyrmion-like microstructures are observed in the room temperature range.

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Conflict of interest

The authors declare that they have no conflict of interest.

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