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Electronic structure and quantum spin fluctuations at the magnetic phase transition in MnSi☆

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ABSTRACT

The effect of spin fluctuations on the heat capacity and homogeneous magnetic susceptibility of the chiral magnetic MnSi in the vicinity of magnetic transition has been investigated by using the free energy functional of the coupled electron and spin subsystems and taking into account the Dzyaloshinsky-Moriya interaction. For helical ferromagnetic ordering, we found that zero-point fluctuations of the spin density are large and comparable with fluctuations of the non-uniform magnetization. The amplitude of zero-point spin fluctuations shows a sharp decrease in the region of the magnetic phase transition. It is shown that sharp decrease of the amplitude of the quantum spin fluctuations results in the lambda-like maxima of the heat capacity and the homogeneous magnetic susceptibility. Above the temperature of the lambda anomaly, the spin correlation radius becomes less than the period of the helical structure and chiral fluctuations of the local magnetization appear. It is shown that formation of a "shoulder" on the temperature dependence of the heat capacity is due to disappearance of the local magnetization. Our finding allows to explain the experimentally observed features of the magnetic phase transition of MnSi as a result of the crossover of quantum and thermodynamic phase transitions.

1. Introduction

The strongly correlated compound MnSi belongs to the structural type B20 with the space group $P2_13$, for which the absence of an inversion centre is typical [1,2]. Such symmetry feature causes the appearance of the band gap in the electronic spectrum [3], and leads to the appearance of the antisymmetric relativistic Dzyaloshinskii-Moriya (DM) exchange [4,5]. In Ref. [4]. it was proposed that the Ginzburg-Landau functional describes the competition of exchange, anisotropic, and DM-interactions, which results in the formation of a long-period helical ordering with a fixed direction of a spin superstructure wave vector (ferromagnetic helicoid) observed in neutron diffraction studies [5]. Analysis of the Bak-Jensen model [4] basing on the renormalization group showed the possibility of formation a helicoidal ferromagnetic phase with a wave vector, the magnitude of which is proportional to a DM parameter and inversely proportional to an inhomogeneous part of the exchange interaction. The ferromagnetic helicoidal state within the framework of the Bak-Jensen model was obtained also using the Fermi-liquid approach [6] and it was shown that in the absence of the DM-interaction, the model describes a weak ferromagnetic state.

The ferromagnetic ground state for the crystal structure of MnSi was considered in the framework of the ab initio calculations [3]. A number of solutions were obtained, which differ by the values of the Hubbard's interaction parameters, form of the density of states, and the values of the magnetic moments. In the range of the Hubbard parameter values: $0 < U < 4$ eV, values of the magnetic moments are approximately 2.5 times larger than follows from the experiment [7]. For $U > 4$ eV the calculated values of the magnetic moments in dependence on the values of the Hubbard's interaction parameters sharply decrease and become approximately two times lower than the experimentally observed [7]. To choose the most suitable model of the electronic structure, a more detailed comparison with the experimental data is needed, in particular, of the electronic heat capacity and magnetic susceptibility.

Difficulty of the correct theoretical description of the magnetic state of MnSi follows also from the experimental data on the phase transition, whose nature has not been definitely ascertained. Renormalization group analysis [4] leads to the conclusion that the observed magnetic transition is not the second-order transition. On the other hand, according to the experiment close to the temperature of the magnetic transition T_C, the lambda-like maxima are formed on temperature dependencies of the heat capacity ($C(T)$) and homogeneous magnetic

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susceptibility of MnSi. However, in the interval $T_S > T > T_C$ there is a "shoulder" on the $C(T)$ dependence [8]. According to results of the small-angle neutron scattering [9,10], chiral fluctuations of the helicoidal structure are observed in transition under consideration. Whereas the radius of spin correlations close to the temperature T_S is approximately equal to the period of the helicoidal superstructure, it then drops sharply with the temperature growth.

In the work [11], the theory of a prolonged first-order phase transition was developed, based on the Ginzburg-Landau functional described by the interaction of fluctuation modes [11]. At that, summands of the intermode interaction were considered without accounting of the specific electronic structure of MnSi and in the lowest fourth order in powers of the magnetization. Moreover, chiral fluctuations observed experimentally, as well as the "shoulder" on the temperature dependence of the heat capacity, were not obtained in these calculations [10,11].

In the present work, we develop a phenomenological approach to the description of phase transitions in helicoidal ferromagnets. The quantum and thermal fluctuations of the electron density are considered within the model of the ground ferromagnetic state of MnSi, supplemented with account for the DM-interaction. Evolution of the electron and spin structures is studied in the temperature vicinity of the magnetic phase transition, as well as the experimental features of the temperature dependences of the heat capacity and homogeneous magnetic susceptibility are explained.

2. Model

The Hamiltonian of the Hubbard-Kanamori model [12] for the ferromagnetic state of strongly correlated electrons, supplemented by addends, which take into account electron density fluctuations, associated with intra-atomic Hubbard and Hund interactions, has the form

$$H = H_0 + \delta H_U. \tag{1}$$

Here $H_0 = \sum_{\mathbf{k},m,\sigma} \varepsilon^{(0)}_{\mathbf{k},m,\sigma} a^+_{\mathbf{k},m,\sigma} a_{\mathbf{k},m,\sigma}$ is Hamiltonian of d-electrons, which was diagonalized in one of the methods, based on the mean-field approximation; $a^+_{\mathbf{k},m,\sigma}(a_{\mathbf{k},m,\sigma})$ is electron creation (annihilation) operator; $\mathbf{k}$ is vector of quasi-momentum, m is index of d-orbital (see the work [3] and references in it); $\sigma(=\pm 1)$ is spin index; $\varepsilon^{(0)}_{\mathbf{k},m,\sigma} = \varepsilon^{(0)}_{\mathbf{k},m} - \sigma\Delta$ is electronic spectrum of d-electrons calculated in the mean-field approximation; Δ is spin splitting of electron energies (independent of m [3]);

$$\delta H_U = \frac{1}{4}\sum_q \left[(U - J/2)|\delta n_q|^2 - (U+J)\sum_m |\delta n_{q,m}|^2 \right] - \sum_q \left[J|\delta S^{(z)}_q|^2 + (U-J)\sum_m |\delta S^{(z)}_{q,m}|^2 \right] \tag{2}$$

is the additive to the mean-field approximation, which takes into account multiparticle correlations (U and J are the parameters of Hubbard and Hund interactions); $\delta n_{\mathbf{q}} = \sum_m \delta n_{\mathbf{q}m}$, $\delta S = \sum_m \delta S^{(z)}_{\mathbf{q},m}$, $\delta n_{\mathbf{q},m} = \sum_\sigma n_{\mathbf{q},m,\sigma} - \delta_{q,0} N^{(0)}_m$, $n_{\mathbf{q},m,\sigma} = a^+_{\mathbf{k},m,\sigma} a_{\mathbf{k}+\mathbf{q},m,\sigma}$, $\delta S^{(z)}_{\mathbf{q},m} = S^{(z)}_{\mathbf{q}m} - \delta_{q,0} M^{(0)}_m$, $S^{(z)}_{\mathbf{q}m} = \sum_\sigma \sigma n_{\mathbf{q},m,\sigma}/2$, while $M^{(0)}_m$ and $N^{(0)}_m$ are the average values of the spin and charge density operators in the mean-field approximation.

Turning to the determination of the partition function, let us use the Matsubara representation of the interaction $(H(\tau) = \exp(-\tau H_0) H \exp(\tau H_0))$,

$$Z = Sp T_\tau \exp\left(-\int_0^\beta H(\tau) d\tau \right), \tag{3}$$

where T_τ is ordering operator in the Matsubara time τ.

Performing the Fourier transformations in (3) and introducing the 4-vector, we average over all directions of the spin axis of quantization. For this, we introduce the unit vectors $\mathbf{e}^{(1)}_{q,m}$ and $\mathbf{e}^{(2)}_{q,m}$ whose directions will be averaged.

$\mathrm{Re}\mathbf{S}_{q,m} = (\mathrm{Re}S^{(z)}_{q,m})\mathbf{e}^{(1)}_{q,m}$, $\mathrm{Im}\mathbf{S}_{q,m} = (\mathrm{Im}S^{(z)}_{q,m})\mathbf{e}^{(2)}_{q,m} (\mathbf{e}^{(j)}_{q,m} = \mathbf{e}^{(j)}_{-q,m}, j = 1, 2)$

Accounting of the electron density fluctuations is carried out using the formalism of Stratonovich - Hubbard transformations [13], which reduces multiparticle interactions in (2) to the interaction of electrons with fluctuating exchange ($\boldsymbol{\xi}$) and charge (η) fields.

$$Z = \int_{-\infty}^{\infty} (d\boldsymbol{\xi}, d\eta) \exp(-\Phi(\boldsymbol{\xi}, \eta)/T) \exp\left\{ -\sum_{q,m} \left|\eta_{q,m}\right|^2 - b\left|\sum_{q,m} \eta_{q,l,m}\right|^2 - (ic/T)\sum_m \eta_{0,m} N^{(0)}_m \right\} \times \exp\left\{ -a\left|\sum_{q,m} \boldsymbol{\xi}_{q,m}\right|^2 - \sum_{q,m} \left|\boldsymbol{\xi}_{q,m}\right|^2 + 2(c/T)\sum_m (M^{(0)}_m - \Delta/U)\xi_{0,m,z} \right\} \tag{4}$$

where: $a = JU(U-J)^{-1}(U+5J)^{-1}$, $b = 4U(U-5J)^{-1}$, $c = (UT)^{1/2}$, $\Phi(\boldsymbol{\xi}, \eta) = -T \ln Sp T_\tau \exp(-H_{eff}(\boldsymbol{\xi}, \eta)/T)$ is the free energy functional of electrons, moving in one of the configurations of stochastic exchange ($\boldsymbol{\xi}$) and charge (η) fields,

$$H_{eff} = \sum_{k,m,\sigma} \varepsilon^{(0)}_{\mathbf{k},m} a^+_{k,m,\sigma} a_{k,m,\sigma} + c\sum_{\nu,m} (\boldsymbol{\xi}_{\nu,m}\mathbf{S}_{\nu,m} + (i/2)\eta_{\nu,m} n_{\nu,m}) \tag{5}$$

is effective Hamiltonian, $\mathbf{S}_{\mathbf{q}} = \sum_{\sigma,\sigma'} a^+_{\mathbf{k},m,\sigma} \boldsymbol{\sigma}_{\sigma,\sigma'} a^+_{\mathbf{k}+\mathbf{q},m,\sigma'}$ is Fourier transform of the spin density operator on the site, $\boldsymbol{\sigma}_{\sigma,\sigma'}$ is the vector of Pauli matrices $(d\eta d\boldsymbol{\xi}) = \left[\frac{U-J}{(U+N_fJ)}\right]\prod_m d\boldsymbol{\xi}_{0,m} d\eta_{0,m} \left[\prod_{q\neq 0,j}\left(\frac{U-J}{(U+N_fJ)}\right)\prod_m d\boldsymbol{\xi}^{(j)}_{q,m} d\eta^{(j)}_{q,m}\right]$, j is index, numbering real and imaginary parts of the ξ-field vector: $\boldsymbol{\xi}_{q,m,\gamma} = \boldsymbol{\xi}^{(1)}_{q,m,\gamma} + i\boldsymbol{\xi}^{(2)}_{q,m,\gamma}$, $\sum_\nu (\ldots) = T\int_0^{T^{-1}} d\tau \sum_{\boldsymbol{\nu}} (\ldots)$.

To describe the ferromagnetic helicoidal ordering, the expression obtained for the free energy of the ferromagnetic state should be corrected in order to describe the energy of the Dzyaloshinskii-Moriya interaction in the mean-field approximation. For this purpose, we will perform the following substitutions[1]

$$H_{eff} \to H_{eff} - \sum_m \left[\mathbf{h}^{(D)}_{\mathbf{q}_0,m} \times \mathbf{S}_{-\mathbf{q}_0,m}\right] \text{ and then } \boldsymbol{\xi}_{q,m} = \boldsymbol{\xi}_{q,m} - \mathbf{h}^{(D)}_{\mathbf{q}_0,m}/c. \tag{6}$$

Here $\mathbf{h}^{(D)}_{\mathbf{q}_0} = \left[\mathbf{M}_{\mathbf{q}_0,m} \times \mathbf{d}_{-\mathbf{q}_0}\right]$ is the Dzyaloshinsky's mean field, $\mathbf{d}_{\mathbf{q}_0} = id\mathbf{q}_0$, d is the Dzyaloshinskii constant, $\mathbf{M}_{\mathbf{q}_0,m}(= \langle \mathbf{S}_{\mathbf{q}_0,m}\rangle)$ is vector of inhomogeneous magnetization on the vector $\mathbf{q}_0$.

Here and below we take into account the fact that the first-principle electronic structure of MnSi does not depend on the quantum number m for $|\varepsilon - \mu| \le UM^{(0)} \sim 1$eV [3]. In this case, the chemical potential of the electronic system is located in the lower band, with orbital degeneracy equal to four [3].

3. The calculation of the statistical sum for considered problem of chiral magnetics

Quantum-statistical calculation of the expression for the statistical sum in the considered problem of chiral magnetics with anomalously large periods of the magnetic spin structure will be carried out on the basis of the approximation of homogeneous local fields [14]. In this case, the spatial-temporal inhomogeneity of the spin system was described by taking into account ($\mathbf{q}$, ω)-dependence of the vertex part of the second order - $\phi^{(2)}(q, q') = \delta_{q,q}\chi^{(0)}_q$, which coincides with the paramagnetic dynamic Pauli susceptibility. The latter determines the dynamic exchange gain factor, anomalously strongly dependent upon $\mathbf{q}$ and ω in ferromagnetic and long-period spin structures and near T_C. In the framework (see, for example [15],) of the well-known approximation of the Lindhard function ($\chi^{(0)}(\mathbf{q}, \omega)$), we have

[1] This replacement turns out to be equivalent to adding to the free energy functional the term $\mathbf{d}_{\mathbf{q}_0}[\mathbf{M}_{\mathbf{q}_0} \times \mathbf{M}_{-\mathbf{q}_0}]$.