PHYSICS OF OUR DAYS

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Helical itinerant MnSi magnet: magnetic phase transition

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Contents

Abstract. **New studies of the phase transition and phase diagram of the chiral MnSi magnet are reported. New results are obtained in the course of analysis of experimental data on heat capacity, thermal expansion, elastic properties, electrical resistance, neutron scattering, and theoretical modeling.**

Keywords: helical magnet, magnetic phase diagram, phase transitions

1. Introduction

MnSi — a helical itinerant magnet — is crystallized in the structure type B20 belonging to the space group $P2_13$, which has no center of symmetry. Although it has been studied intensively for several decades, this magnetic material still remains at the center of attention of many researchers. This circumstance is due to a whole series of reasons, some of which are as follows.

(1) MnSi is an example of a substance with a helical magnetic structure caused by the Dzyaloshinskii–Moriya interaction [1].

(2) Studies of the physical properties of MnSi at high pressures have revealed a number of intriguing features, such as a quantum phase transition [1, 2], non-Fermi-liquid behavior [3, 4], and 'partial' helical order [5], which still await further study.

(3) The so-called phase A, appearing in MnSi in a magnetic field, which was identified as a skyrmion crystal [1, 6], has proven to be extremely sensitive to various kinds of actions and is considered a promising material for spintronics [7].

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(4) The phase transition in MnSi at a temperature of 29 K and normal pressure from the paramagnetic state into the helical state has finally been acknowledged as a first-order phase transition [8], which casts doubts upon early assertions about the shape of the phase diagram of MnSi at high pressures [9].

(5) Finally, which is of great importance, MnSi constitutes a simple binary compound synthesized from elements that allow deep purification. MnSi possesses a comparatively low temperature of congruent melting ($\sim$ 1500 K), which facilitates growing large crystals. To date, large and sufficiently perfect MnSi single crystals of high purity have been grown in a number of laboratories, and thus have become accessible for diverse physical studies.

This article comprises a survey of new data obtained in the course of studies on heat capacity, thermal expansion, elastic properties, electrical resistance, and neutron scattering in MnSi, in particular, at high pressures and in strong magnetic fields.

2. Magnetic phase transition in MnSi

Let us first examine the magnetic phase transition in MnSi at atmospheric pressure in a zero magnetic field. The analysis of a phase transition in any system, as a rule, begins from the mean-field approximation, which disregards fluctuations. The most popular approximation is the phenomenological Landau model based on symmetry considerations, in which the expansion of the thermodynamic potential Φ in powers of the order parameter η is used. In the simplest case, the Landau expansion is written out as follows:

$$\Phi(P, T, \eta) = \Phi_0 + A\eta^2 + C\eta^3 + B\eta^4 + \dots, \qquad (1)$$

where the coefficients A, B, and C are functions of pressure P and temperature T. In view of the symmetry relative to time reversal, $C \equiv 0$ in the case of magnetic phase transitions, and relationship (1) takes on the following form:

$$\Phi(P, T, \eta) = \Phi_0 + A\eta^2 + B\eta^4 + \dots . \qquad (2)$$

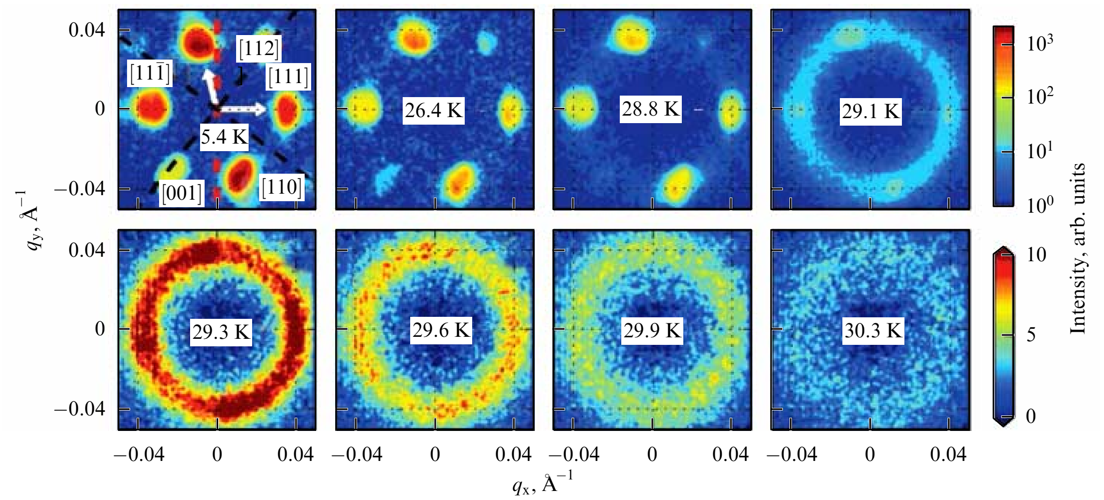

Figure 1. (Color online.) Helicomagnetic phase transition in MnSi at 29 K according to data from neutron experiments [16]; q_x and q_y are the coordinates in the momentum space. It can be seen that, during a phase transition in the temperature interval of 28.8–29.1 K, the discrete Bragg peaks corresponding to the helical magnetic order, are replaced by diffuse magnetic scattering, which is concentrated on the surface of a sphere in the reciprocal space. These effects reflect the existence of helical magnetic fluctuations in the immediate proximity to the temperature of the phase transition.

At the point of the second-order phase transition, one has $A = a(T - T_c) = 0$ and the coefficient $B > 0$. For $B < 0$, a first-order phase transition takes place in the system. In this case, a sixth-order term $D\eta^6$ with $D > 0$ should be added to expansion (2) to stabilize the system. The available experimental data indicate a certain lability of the coefficient B in different systems; as a result, the phase transition can alter its nature with a change in the ambient conditions with the appearance of the so-called tricritical point [10].

There are internal reasons for which some phase transitions are first-order in spite of the absence of symmetry limitations. First, there are striction effects, which can arise in real compressible lattices [11, 12]. Competition among two or more order parameters caused by different interactions, if accompanied by strong fluctuations, can also lead to a first-order phase transition. In essence, these factors renormalize the coefficient B and even lead to the appearance of nonzero third-order terms (see the Halperin–Lubensky–Ma effect [13]). No one should be embarrassed by the fact that we are discussing here the evolution of the Landau expansion coefficients, which are the mean-field parameters, together with the fluctuation effects, which seemingly violate the mean-field picture of the phase transition. In reality, the fluctuations are concentrated in a relatively narrow temperature range determined by the Levanyuk–Ginzburg criterion [10], and beyond this range the mean-field picture remains valid; therefore, we can consider fluctuations as a factor that can affect both the sign and the value of the coefficients of the Landau expansion.

Further on, turning to the subject of our study it should be noted that the fluctuational nature of the first-order phase transition in MnSi was indicated for the first time by Bak and Jensen [14], who carried out relevant calculations, apparently influenced by the work of Hansen [15], who revealed an abrupt change in the intensity of the line of superlattice scattering at the point of the phase transition. By the way, Hansen's work was not published in the contemporary scientific literature and remained unknown, until it was mentioned as a reference cited in paper [16].

At present, the fluctuational model of the phase transition in MnSi, based on Brazovskii's theory [17], is becoming popular; here, the role of fluctuations in systems with a fluctuation spectrum possessing a low absolute minimum with a nontrivial value of the moment is emphasized. The authors of Ref. [16] have analyzed the experimental data on small-angle neutron scattering in the vicinity of the phase transition in MnSi and developed a theory which, in their opinion, describes well the totality of the experimental data (Fig. 1).

It is necessary to recall that an important feature of the magnetic phase transition in MnSi is the presence of a secondary gently sloping maximum or a shoulder in the curves of the heat capacity, thermal expansion, thermal resistance, and sound absorption, as is evident, for example, from Fig. 2. The shoulder is located in the existence domain boundaries of the paramagnetic phase but is characterized by powerful chiral fluctuations, as follows from the data on neutron scattering [18]. According to the authors of paper [16], they obtained a successful description of this feature of the phase transition in MnSi, although their graphic proof of the existence of a shoulder (see Fig. 2) is by no means convincing.

3. Vollhardt 'invariant'

Let us say several words about the so-called Vollhardt invariant. In 1997, D Vollhardt revealed that the curves of the heat capacity of a number of highly correlated systems measured at different values of the thermodynamic parameters (for example, pressure, magnetic field, etc.) intersect at almost the same temperature [19]. This effect is distinctly visible in the curves of heat capacity, thermal expansion, elastic moduli, and sound absorption (Fig. 3) [20]. The very term 'Vollhardt invariant' was apparently first used in Ref. [21], where the data on the heat capacity of MnSi, Mn(Co)Si, and Mn(Fe)Si were analyzed.

The presence of this invariant is connected with the existence of a specific characteristic energy determining the population of helical magnetic fluctuations, which naturally indicates the occurrence of the Dzyaloshinskii–Moriya interaction [21]. This conclusion is supported in Ref. [16], where it is asserted that the appearance of the Vollhardt

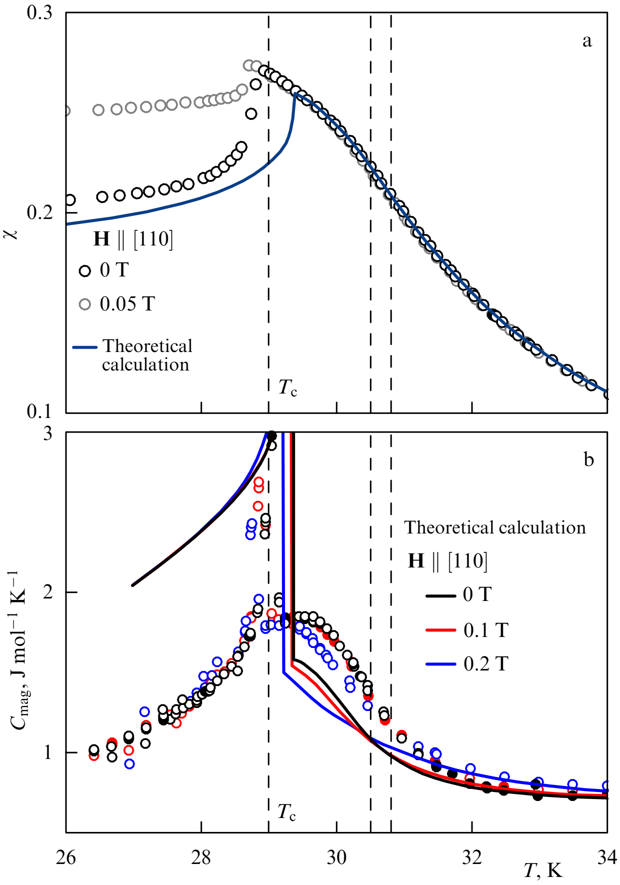

Figure 2. (Color online.) Comparison of the results of theoretical calculations based on the Brazovskii model [16] and experimental data, which characterize (a) the magnetic susceptibility χ, and (b) the heat capacity C_{mag} of MnSi according to the data from Ref. [16]. The solid curves correspond to the theoretical calculations; the circles show experimental data.

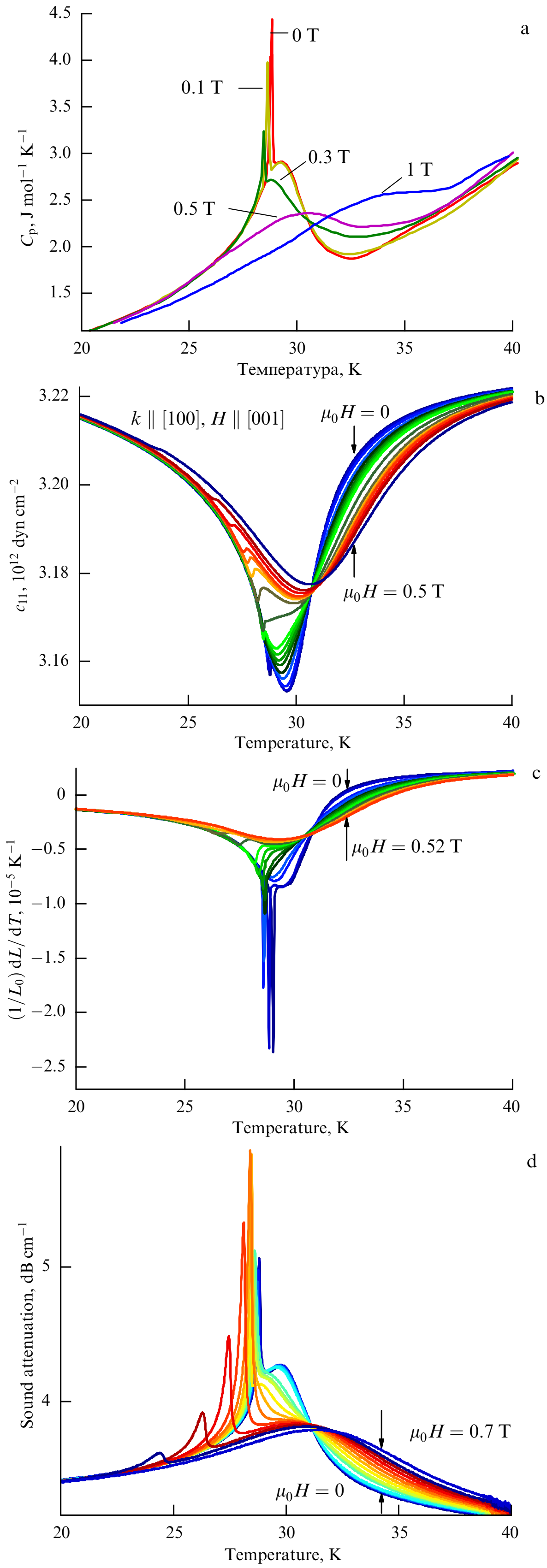

Figure 3. (Color online.) (a) Heat capacity, (b) elastic modulus c_{11}, (c) coefficient of thermal expansion, and (d) coefficient of sound absorption as functions of temperature and magnetic field in a phase transition in MnSi [20]; μ_0 is the permeability of vacuum.

invariant directly follows from the Brazovskii model [17]. A simulation of the situation with the aid of Gaussian functions performed in Ref. [20] shows that the appearance of a pseudoinvariant temperature should always be expected when a magnetic or some other field broadens the appropriate maxima and decreases their amplitudes in such a manner that their integral values remain unaltered (Fig. 4).

Thus, in Ref. [20] we reached the conclusion that the 'point of intersection' or the Vollhardt invariant cannot serve as an indicator of the existence of some specific energy, and the very 'invariant' in general simply represents an approximate point of the intersection of the corresponding curves rather than an invariant, as can be seen from Fig. 3. The very maxima or minima, illustrated in Fig. 5, are identified in Ref. [20] as smeared phase transitions.

4. Phase transition in MnSi according to simulations by the Monte Carlo method

Here, we should pay special attention to paper [22], which is devoted to an analysis of the properties of a three-dimensional lattice system of spins with the aid of the classical Monte Carlo method. Together with the ferromagnetic exchange interaction (J), the authors of Ref. [22] take into account the anisotropic Dzyaloshinskii–Moriya interaction (D). Figure 6, borrowed from Ref. [22], demonstrates an excellent agreement of the results of simulations with the

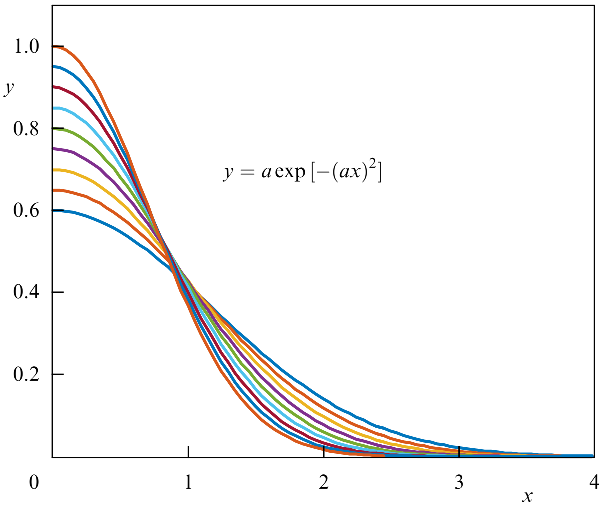

Figure 4. (Color online.) Simulation of the effect of the Volkhardt intersection with the aid of a Gaussian function and a variation of the width a [20].

experimental data for MnSi in a zero magnetic field. In Ref. [23], a study of the same system of spins was carried out by the same numerical method but with a variable ratio between the exchange interaction and the Dzyaloshinskii–Moriya interaction (J/D). The use of the J/D ratio should not be misleading, since the terms containing J and D enter into the Hamiltonian as summands, and the zeroing of the corresponding terms does not lead to divergence.

As it turns out, the shoulder in the curve of the heat capacity of the model system of spins appears as a result of a perturbation of the ferromagnetic second-order phase transition by helical fluctuations caused by the Dzyaloshinskii–Moriya interaction. With an increase in the contribution of this interaction, a first-order phase transition occurs in the system, and at $J/D \approx 1$ the behavior of the heat capacity of the system becomes similar to that experimentally observed in the cases of MnSi and Cu_2OSeO_3 (see Section 6): a sharp peak appears, which corresponds to a first-order transition, as does a gently sloping maximum or a shoulder at temperatures that somewhat exceed the temperature of the phase transition (Fig. 3).

As follows from the calculations carried out, the observed maxima in the temperature dependences of the physical quantities (Fig. 7) are connected with the 'smeared' second-order phase transition, which is in complete agreement with the conclusion made in Ref. [20]. However, it was assumed in Ref. [20] that the smearing is connected with a natural imperfection of the MnSi crystal, whereas it follows from the calculations performed by the Monte Carlo method that the degradation of the ferromagnetic second-order phase transition is a result of helical fluctuations.

Thus, the region of the maximum possesses a complex structure, which corresponds to the interaction of two fluctuating order parameters; as a result, the system cannot pass into the ordered state continuously, but does so jumpwise, via a first-order transition.

5. Phase transition in MnSi in a magnetic field

Let us now turn to studies of the phase transition in MnSi in a magnetic field. In Ref. [24], detailed measurements of the heat capacity of MnSi in magnetic fields were carried out, which revealed a nontrivial behavior of the heat capacity in the vicinity of the phase transition (Fig. 8). The authors of Ref. [24] assume that the results presented in Fig. 8 indicate the existence of a tricritical point at a temperature of 28.5 K and a magnetic field of 340 mT (Fig. 9). Simultaneously, the authors emphasize that the observed anomalies in the behavior of heat capacity at the boundaries of the skyrmion phase indicate their thermodynamic nature.

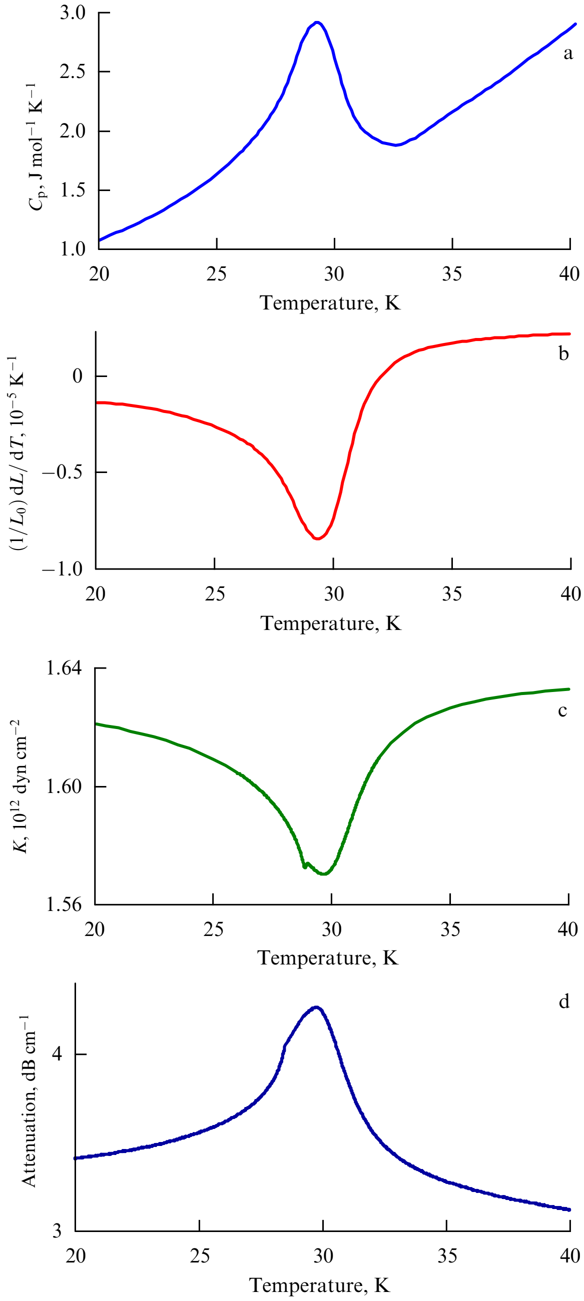

Figure 5. (a) Heat capacity, (b) coefficient of thermal expansion, (c) bulk modulus K, and (d) coefficient of sound absorption as functions of temperature in a phase transition in MnSi [20]. The sharp peaks caused by the first-order phase transition are removed to more distinctly show the anomalies, which, apparently, are characteristic of the smeared phase transition.

The results of ultrasonic studies of the phase diagram of MnSi in a magnetic field are presented in Figs 10 and 11 [25]. Notice that the studied MnSi sample had the shape of a disk, which affected the value of the demagnetization factor at

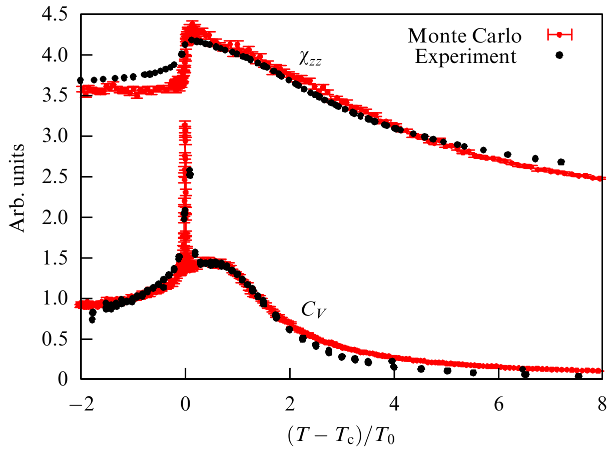

Figure 6. (Color online.) Heat capacity and the magnetic susceptibility in the phase transition in a three-dimensional system of Heisenberg spins with the Dzyaloshinskii–Moriya interaction according to data calculated by the Monte Carlo method [22]. Experimental data for MnSi were borrowed from Ref. [16].

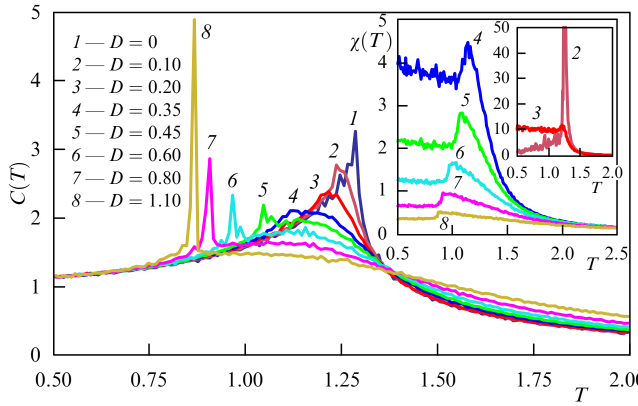

Figure 7. (Color online.) Behavior of the heat capacity at the phase transition in a three-dimensional system of Heisenberg spins with the Dzyaloshinskii–Moriya interaction with a variation in the Dzyaloshinskii–Moriya coupling constant D and the exchange interaction constant $J = 1$ according to data of simulations by the Monte Carlo method [23]. Temperature is given in units of J; the heat capacity, in dimensionless units. In the insets, the magnetic susceptibility $\chi(T)$ is shown at different values of D.

various orientations of the sample in the magnetic field. As a result, the magnetic scale of the corresponding dependences proves to be somewhat different (see Figs 10 and 11). Notice also that the higher the uniformity of the magnetic field in the sample, the less the domain of existence of the phase of the

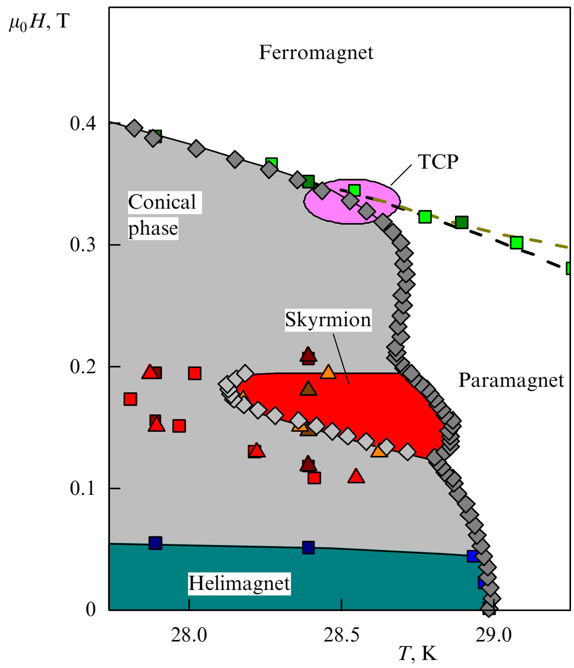

Figure 9. (Color online.) Magnetic phase diagram of MnSi. The oval contour outlines the region of the location of the supposed tricritical point (TCP) [24].

skyrmion crystal (in the $\mathbf{k} \perp \mathbf{H}$ configuration, the magnetic field is directed parallel to the plane of the disk and is distributed more uniformly than in the case of $\mathbf{k} \parallel \mathbf{H}$).

As can be seen from Fig. 10, an abrupt change in the elastic moduli c_{11} and c_{33} in the magnetic phase transition in MnSi upon the imposition of a magnetic field first diminishes rapidly, reaching practically zero values in the domain of existence of the skyrmion phase, then grows in the region of magnetic fields and temperatures indicated in Ref. [24] as the tricritical coordinates, and further decreases to negligibly small values. All this is illustrated in Fig. 12, which demonstrates the dependence of the jumps of the moduli and the amplitude of the attenuation factor on the magnetic field at different orientations. Let us emphasize that at the tricritical point a divergence is expected of heat capacity and compressibility, and, therefore, of such quantities as $1/c_{ij}$ [10]. However, nothing similar is observed in Figs 10 and 12. Nor is divergence observed in Fig. 8. Nevertheless, a specific anomaly, which manifests itself in the appearance of maximum jumps of elastic constants and of the coefficient of ultrasound absorption, is observed in the 0.3–0.4 T region of a

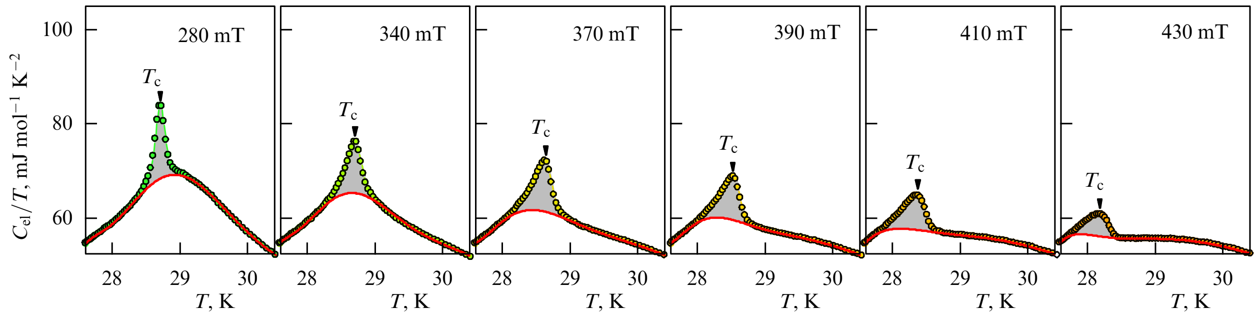

Figure 8. Heat capacity divided by the temperature at the phase transition in MnSi depending on the magnetic field [24]. According to the assumption of the authors of Ref. [24], a tricritical transition takes place in a magnetic field of 340 mT.

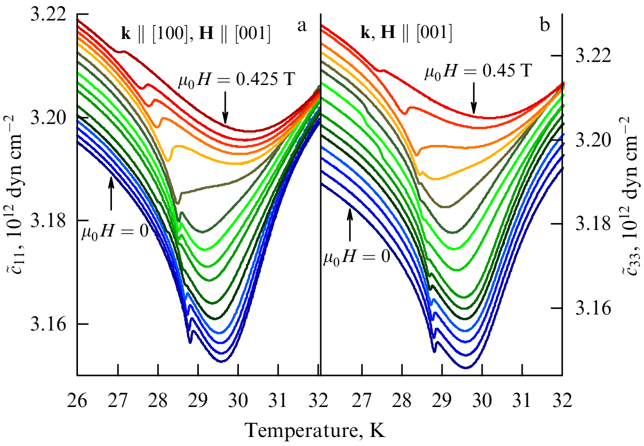

Figure 10. (Color online.) Temperature dependence of the elastic moduli $\tilde{c}_{11}$ and $\tilde{c}_{33}$ in the region of the phase transition in MnSi upon a variation in the magnetic field value. The tilde sign denotes the measured values corresponding to a cubic crystal with a tetragonal anisotropy induced by the magnetic field, in order to distinguish them from truly 'tetragonal' values [25]; k is the wave vector. The curves are shifted relative to each other along the ordinate axis.

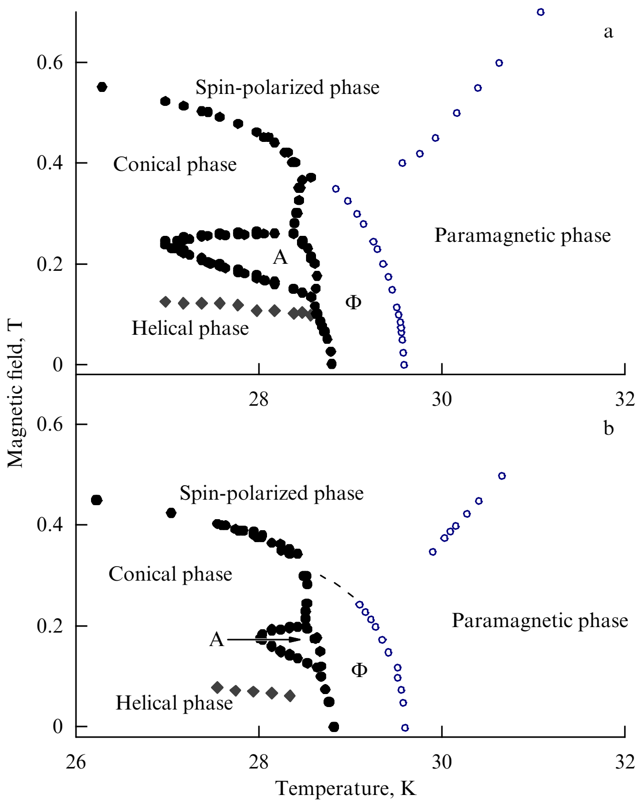

Figure 11. Magnetic phase diagram of MnSi according to Ref. [25]: A, skyrmion crystal; Φ, region of strong helical fluctuations. It can be seen that the domain of the existence of the skyrmion phase depends on the orientation of the sample in the magnetic field.

magnetic field. It is precisely here that the line of the minima of the elastic moduli corresponding to the smeared phase transition touches the line of phase transitions, which makes this region similar to an end critical point, at which the line of second-order phase transitions is joined with the line of the first-order transition [25] (see Fig. 11).

Additional information on the evolution of the magnetic phase transition in MnSi can be extracted from the data on

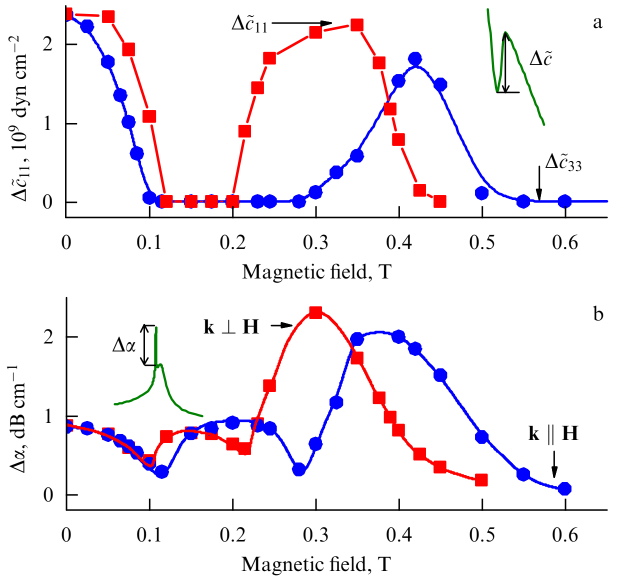

Figure 12. (Color online.) (a) Values of the jumps in elastic moduli, and (b) partial amplitudes of the attenuation coefficients at the phase transition in MnSi as functions of the magnetic field. The difference between the two systems of data given in figures (a) and (b) is connected with the difference among the magnitudes of the demagnetization factor [25].

the thermal expansion [26]. Some results of thermal expansion measurements are shown in Figs 13 and 14. It can be seen that in a magnetic field of 0.48 T the bulk anomalies disappear almost completely, which disagrees with the existence of a tricritical point in the magnetic field of 0.4 T in the case of the occurrence of a second-order phase transition for $\mu_{\mathrm{B}} H > 0.4$ T.

Figure 15 displays the results of measurements of jumps in the sample lengths and in the heights of the peaks of the thermal expansion coefficient in the [100] direction at the phase transition in MnSi at various values of the magnetic field.

It can be seen that the anomaly of the coefficient of thermal expansion decreases with increasing magnetic field (see Figs 14, 15); a pronounced dip in the middle of the range corresponds to the region of the skyrmion phase (see Fig. 15). However, nothing indicates a tricritical behavior of the coefficient of thermal expansion. At the same time, the jump in the thermal expansion, decreasing to very low, but finite values (10^{-7}) with increasing magnetic field suddenly becomes zero. In reality, the jump is simply smeared, so that the measurement of its value becomes impossible. In Ref. [26], we concluded that the phase transition in MnSi is always a first-order transition, whose discontinuous picture is violated by heterophase fluctuations.

6. Phase diagram of MnSi at high pressures

In this section, we discuss the situation with the phase diagram of MnSi at high pressures. In papers [27, 28], Pfleiderer et al. stated, based on the measurements of magnetic susceptibility, that in the curve of the phase transition at a pressure of 1.2 GPa and a temperature of 12 K there is a tricritical point at which the continuous phase transition becomes a first-order transition. This idea found theoretical support [29]. And although the interpretation of the results of the measurements of the magnetic susceptibility

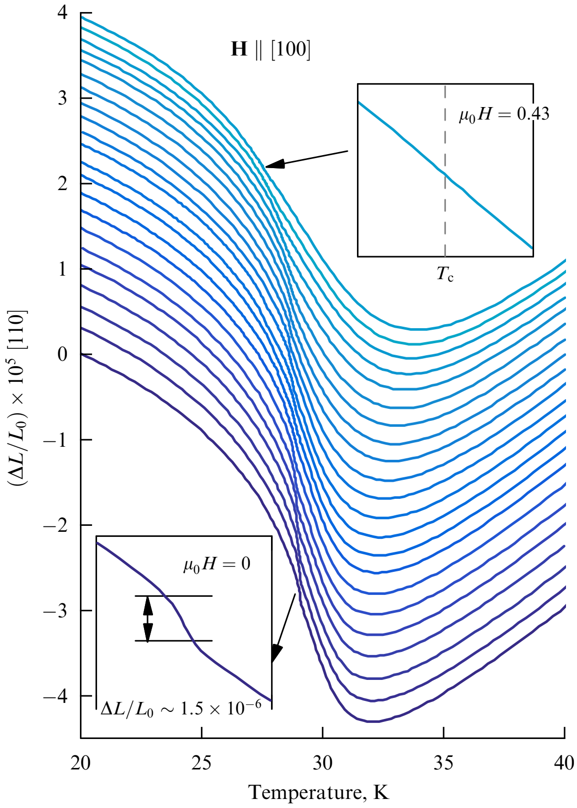

Figure 13. Linear thermal expansion of an MnSi crystal as a function of temperature at various values of a magnetic field. The curves are shifted relative to each other along the ordinate axis. It can be seen that the anomaly connected with the magnetic phase transition disappears in strong magnetic fields [26].

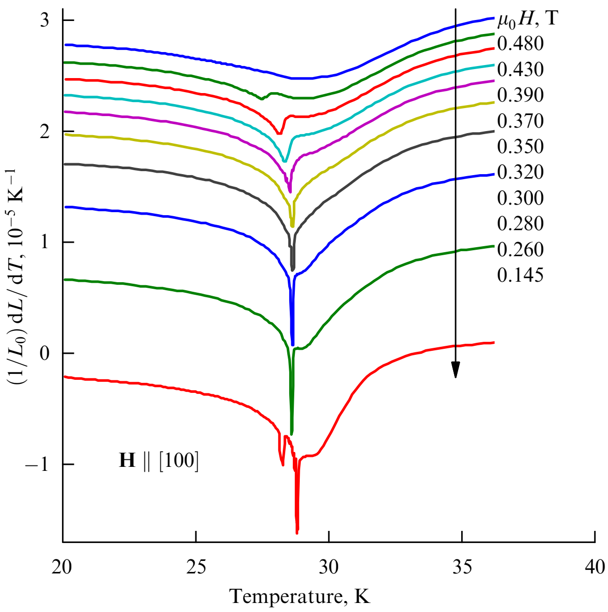

Figure 14. Temperature dependence of the linear thermal expansion coefficient of the MnSi crystal. The curves are shifted relative to each other along the ordinate axis. Degradation and disappearance of peaks of the coefficient of thermal expansion with an increase in the magnetic field are clearly visible. In the lower curve, a peak corresponding to the skyrmion phase is also noticeable [26].

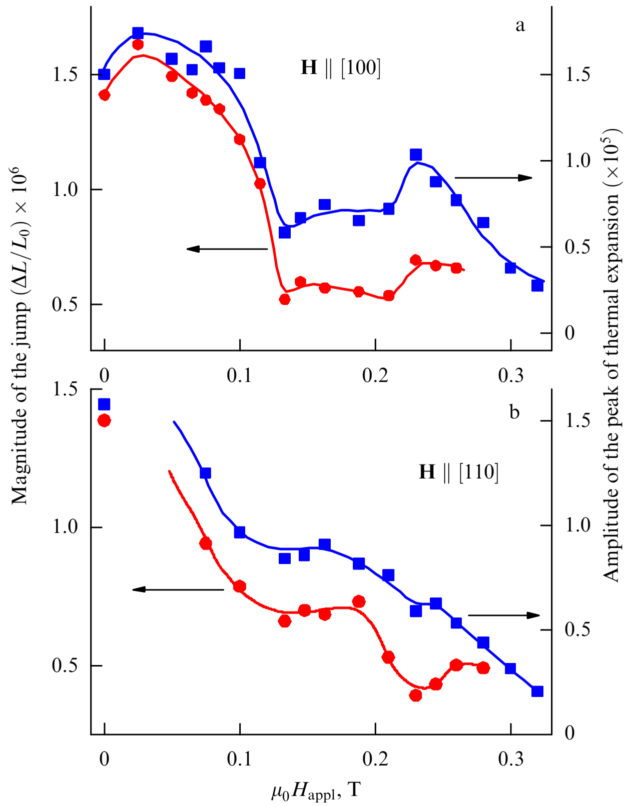

Figure 15. Dependence of the jumps in thermal expansion and values of the peaks of the thermal expansion coefficient of an MnSi crystal on the magnetic field. Both quantities decay with increasing magnetic field. The local anomaly in the middle of the range corresponds to the skyrmion phase. The jumps suddenly effectively decrease to zero in fields of 0.27–0.3 T (see the main text) [26].

of MnSi at high pressures was criticized (see review [1]), the ‘tricritical idea’ continued living. The measurements of bulky effects in the limit of low temperatures seemingly indicated the discontinuous nature of the volume change upon a phase transition in MnSi [30, 31]. In Ref. [32], this problem was analyzed in connection with the results of the measurements of the electrical resistance of MnSi. First of all, notice that the ‘tricritical idea’ was intensely popularized until the phase transition in MnSi was considered as a second-order one. At present, the situation, as we saw above, is fundamentally different, which, however, does not prevent us from examining the issue.

Figure 16 illustrates the dependence of the thermal expansion of MnSi on the temperature at atmospheric pressure. It can be seen that the weak first-order phase transition is hardly noticeable against the background of the extensive bulky anomaly. It is obvious that this transition could not be detected in the relatively rough experiments at high pressures [30, 31]. The authors of Refs [30, 31] apparently observed the bulky anomaly (Fig. 16), which, to a considerable extent, is localized as a result of ‘freezing’ thermal fluctuations at low temperatures and high pressures. This situation is illustrated in Fig. 17, where the isotherms of the electrical resistance of MnSi at different temperatures are plotted. It is clearly seen that the region of anomalous scattering of carriers on magnetic fluctuations shrinks with decreasing temperature and increasing pressure. At temperatures on the order of 2–5 K, the region of the anomalous scattering, one way or another connected with the volume anomaly [20], becomes too narrow to imitate the situation with the smeared first-order transition. However, these reasons do not force the authors of different concepts to

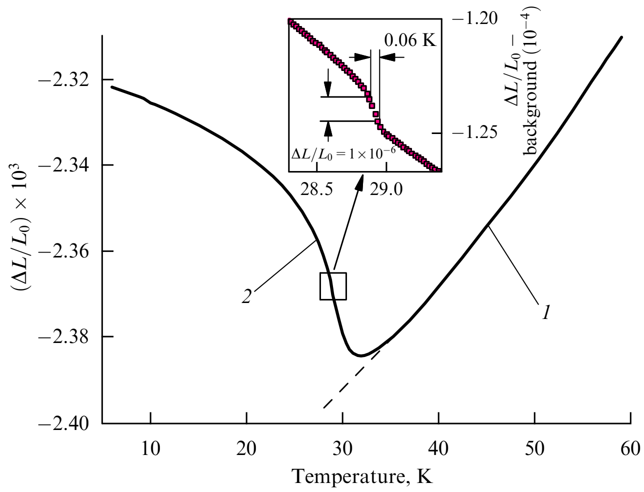

Figure 16. Linear thermal expansion of MnSi, which illustrates the relationship between the bulky anomaly and the first-order phase transition [32].

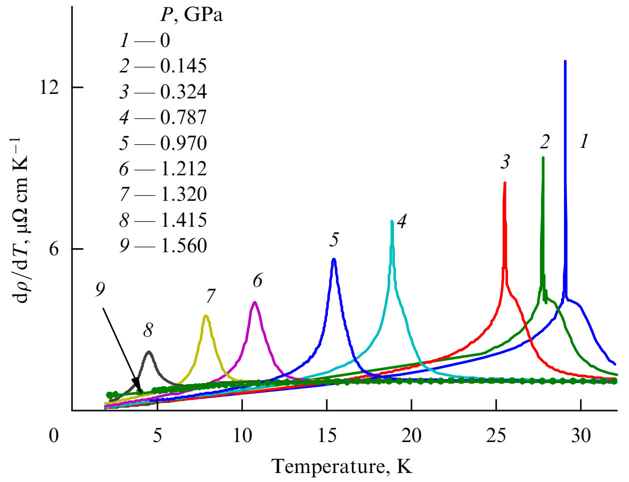

Figure 18. (Color online.) Temperature derivative of the resistivity $d\rho/dT$ at a phase transition in MnSi at high pressures.

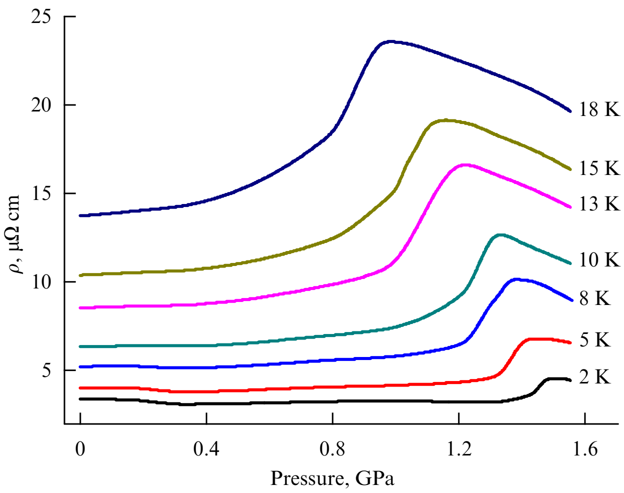

Figure 17. Isotherms of the electrical resistance of MnSi, which demonstrate the evolution of the fluctuation region in the vicinity of the phase transition [32].

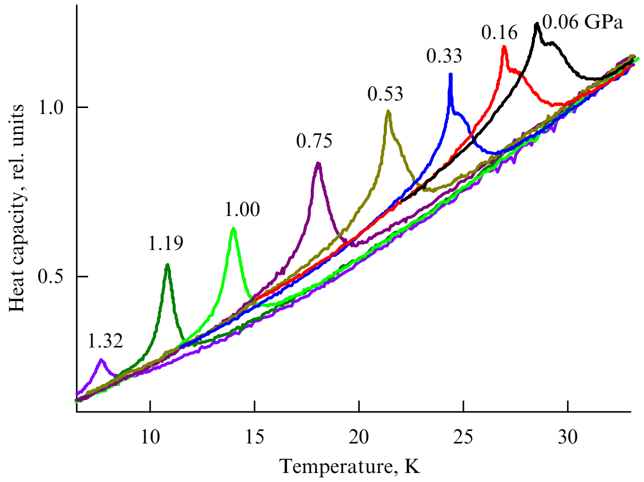

Figure 19. (Color online.) Heat capacity in the vicinity of the phase transition in MnSi at various pressures [33].

reexamine their views. For example, the existence of a tricritical point in the curve of the phase transition in MnSi was discussed in recent review [8], although its authors encountered some difficulties, since they recognize that the phase transition in MnSi is a first-order transition in two limiting cases: at atmospheric pressure and at high pressure and low temperatures. But then, what can we say about the tricritical point? The authors of Ref. [8] suggest a 'Solomon' solution: the tricritical point corresponds to a passage from a weak first-order transition to a strong transition! This suggestion resembles the joke about a dispute between two museums in the USA about which of them possesses the authentic skull of the hero of the Mexican revolution Pancho Villa.[1] The skulls were different in sizes, and the museums came to an agreement that one of the skulls belonged to the young Pancho Villa and the other skull to the adult Pancho Villa.

[1] Three years after Pancho Villa was assassinated, his grave was uncovered and his head was stolen.

Measurements of the electrical resistance [32] and heat capacity [33] at high pressures made it possible to draw specific conclusions about the phase diagram of MnSi (Figs 18, 19) (the temperature derivative of the electrical resistance in the case of phase transitions in magnetic metals behaves analogously to that of the heat capacity [34]). It can be asserted that the explicit signs of the first-order transition in MnSi [the sharp peak and the shoulder (see Fig. 3)] disappear with an increase in the pressure and a decrease in the temperature.

This may be connected with the suppression of thermal fluctuations, if we assume that the first-order transition in MnSi has a fluctuational origin. On the other hand, it cannot be ruled out that the phase transition is simply smeared at low temperatures and high pressures as a result of emerging the nonhydrostatic stresses.

Nevertheless, the results of work [32, 33] apparently indicate the absence of a strong first-order transition in MnSi as $T \to 0$. The phase diagram of MnSi proposed in Refs [32, 33] is shown in Fig. 20.

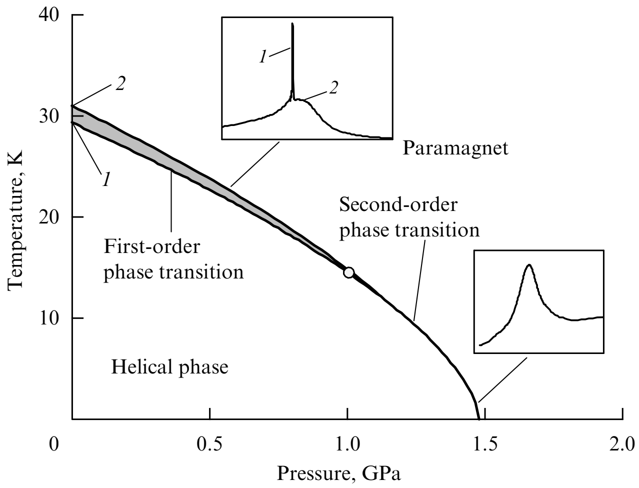

Figure 20. Supposed phase diagram of MnSi at high pressures. The gray region corresponds to the domain of strong helical fluctuations in the paramagnetic phase. The insets illustrate the evolution of the heat capacity and temperature derivative of resistivity $\mathrm{d}\rho/\mathrm{d}T$ depending on pressure. The circle at the beginning of the gray region can correspond to a tricritical point, if the phase transition in MnSi at high pressures is indeed continuous [32].

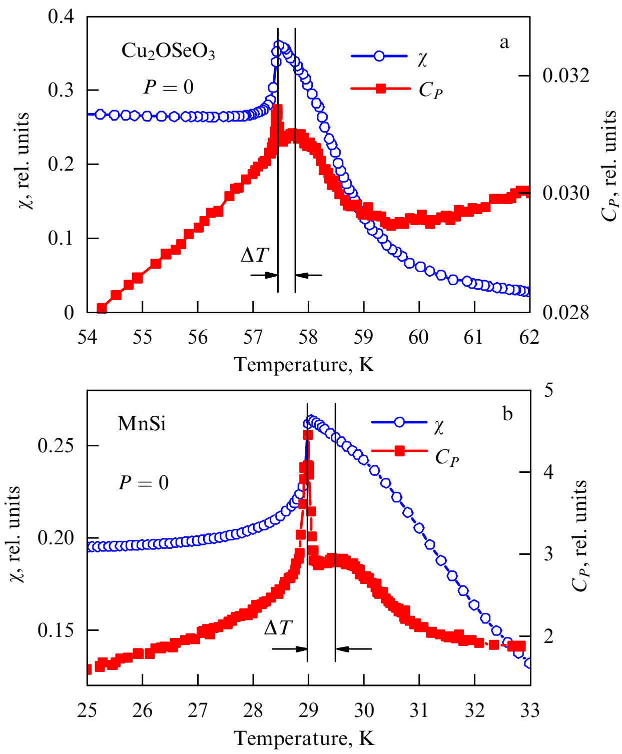

Figure 21. Magnetic susceptibility and heat capacity upon a phase transition in the chiral magnets (a) Cu_2OSeO_3, and (b) MnSi. The similarity between the behavior of the itinerant magnet and the magnet with localized spins is obvious [33].

7. Conclusion

This article is a kind of supplement to our earlier article [1] and does not pretend to an illumination of all achievements and difficult aspects of the problem. As a definite achievement, note the following: at present, it is already universally recognized that the magnetic phase transition in MnSi at atmospheric pressure and zero magnetic field is a first-order phase transition. However, this fact comes into conflict with early presentations of the shape of the phase diagram of MnSi at high pressures and low temperatures and the character of related quantum phenomena [27–29]. Progress in this field requires the development of new experimental techniques for studies at high pressures, which is a question for the future. Nor should we forget the uncommon phenomena that appear upon a phase transition in MnSi in strong magnetic fields [26]. And, finally, let us present Fig. 21 [33], which illustrates the close analogy between the itinerant and Heisenberg magnets, MnSi and Cu_2OSeO_3, respectively, with the Dzyaloshinskii–Moriya interaction, which, apparently, indicates the insignificant role of longitudinal spin fluctuations in the course of magnetic phase transitions [35].

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