Behavior of the electrical resistivity of MnSi at the ferromagnetic phase transition

Alla E. Petrova,[1] E. D. Bauer,[2] Vladimir Krasnorussky,[1] and Sergei M. Stishov[1,2,*]
[1]*Institute for High Pressure Physics of Russian Academy of Sciences, Troitsk, Moscow Region, Russia*
[2]*Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA*
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The itinerant helical ferromagnet MnSi reveals a number of remarkable features, which include tricritical phenomena at the phase transition line, Fermi-liquid breakdown, and so-called partial spin order in the paramagnetic state at high pressures. These features, probably interconnected, so far have no satisfactory explanations though several ideas have been suggested. Some current ideas focus on specifics of the spin fluctuations in the paramagnetic phase of MnSi close to the phase transition line. We report here the results of electrical resistivity measurements of a single crystal of MnSi across its ferromagnetic phase transition line at ambient and high pressures. Contrary to previous work in the field we made use of compressed helium as a pressure medium. Sharp peaks of the temperature coefficient of resistivity characterize the transition line. Analysis of these data shows that at pressures to ~0.35 GPa these peaks have fine structure, revealing a shoulder at ~0.5 K above the peak. That confirms the "abnormal" spin behavior in the narrow region above the Curie point and indicates the existence of a nontrivial fluctuation mode in the paramagnetic phase of MnSi. It is symptomatic that this structure disappears at pressures higher than ~0.35 GPa, which was identified earlier as a tricritical point.

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The intermetallic compound MnSi experiences a second-order phase transition at a temperature T_c slightly below 30 K, acquiring helical magnetic structure and becoming a weak itinerant ferromagnet. On application of pressure the transition temperature T_c decreases and tends to zero at a pressure of about 1.4 GPa.[1] As was noticed in Ref. 2 (see also Ref. 3), a λ-type singularity of the ac magnetic susceptibility χ_{ac} at the phase transition in MnSi deforms gradually with pressure and becomes a simple step at pressures more than 1 GPa. That was grounds to claim the existence of a tricritical point with the coordinates ~1.2 GPa, ~12 K.[2,3] This conclusion was partly disputed in Ref. 4, where measurements of χ_{ac} of MnSi at high pressures, created by compressed helium, were reported. These authors[4] confirmed the existence of a tricritical point on the phase transition line in MnSi but placed it at much lower pressure and at significantly higher temperature ($P_{tr} \cong 0.355$ GPa, $T_{tr} \cong 25.2$ K). Meanwhile new features of MnSi at high pressures were reported. A strong deviation from Fermi-liquid behavior in the paramagnetic phase of MnSi was found in Ref. 5. The unusual spin ordering (partial order) in the paramagnetic phase of MnSi was discovered at high pressure in Ref. 6. Concepts like "blue quantum fog,"[7] "helical spin crystal,"[8] and skyrmionlike spin structures,[9] were proposed in attempts to explain these observations.

Keeping in mind that specifics of the spin fluctuations in the vicinity of the phase transition line may be responsible for the physics observed in MnSi, we have carried out precise resistivity measurements of a MnSi single crystal across the phase transition line at ambient and at high pressures, using a compressed helium technique. The primary purpose was to study the behavior of the temperature coefficient of resistivity $d\rho/dT$ at the transition line. According to the theoretical conclusions,[10–12] the temperature coefficient of resistivity diverges at a second-order magnetic phase transition and can be characterized by a static critical exponent. Contrary to this expectation, we found that the peaks in $d\rho/dT$ at T_c are accompanied by a well-defined shoulder on their high-temperature side, which vanishes when approaching the pressure ~0.35 GPa. This finding nicely correlates with corresponding features in ultrasound attenuation,[13] thermal expansion,[14] and heat capacity,[15] discovered in the critical region of MnSi at ambient pressure, and offers persuasive proof for the existence of fluctuations different from fluctuations of the order parameter, normally occurring in the critical region.

The single crystal of MnSi was grown from a tin flux by dissolving prealloyed Mn and Si in excess Sn. For resistivity measurements, four Pt wires of 25 μm in diameter ere welded to the crystal with dimensions of about $0.5\times0.3\times0.3$ mm^3. The temperature of the magnetic phase transition T_c and the resistivity ratio $R_{300}/R_{(T=2.1)}$, taken at ambient pressure, are equal to 29.25 ± 0.02 K and ≈ 100, respectively. The crystal was placed in a high-pressure cell made of beryllium copper. Fluid and solid helium were used as pressure medium, therefore providing an almost hydrostatic environment for the sample. Temperature was measured by a calibrated Cernox sensor, embedded in the cell body, with an accuracy of about 0.05 K. A calibrated Manganin gauge was used to measure pressure with accuracy about 10^{-3} GPa in the fluid helium domain. In the domain of solid helium, pressure was calculated on the basis of the measured helium-crystallization temperatures and data for the equation of state of helium. The accuracy of pressure measurements in solid helium is estimated as 5×10^{-3} GPa. The resistivity was measured by a four-terminal dc method. The experimental setup, including the high-pressure gas installation and the cryostat, is described in Refs. 4 and 16.

The resistivity measurements of MnSi were carried out along 24 quasi-isobars[17] in the pressure range from zero to 1.5 GPa. Selected experimental data are displayed in Fig. 1. We have tried to describe the resistivity curves in the temperature range from zero to the phase transition region by

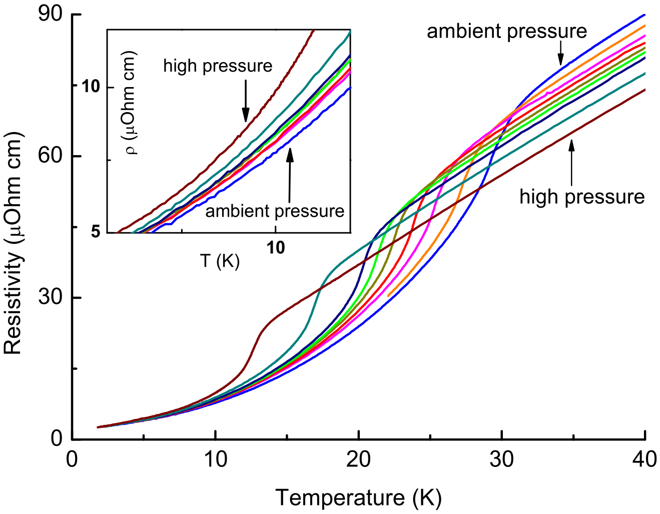

FIG. 1. (Color online) Temperature dependence of the electrical resistivity $\rho(T)$ of MnSi at different pressures. The isobars correspond to pressures 0, 0.2, 0.32, 0.43, 0.54, 0.63, 0.7, 0.885, and 1.13 GPa, counting from the right to the left at the bottom of the figure.

various polynomials that contained potentially important T^2 and/or T^5 terms accounting for scattering by spin and density fluctuations (phonons).[18,19] The overall results appeared to be quite satisfactory though we observed small but systematic deviations of the experimental data points from the corresponding approximations at low temperatures. Replacing the T^2 term with T^n improves the situation but does not correct it entirely, though it always leads to a value of $n<2$. On the other hand, as is seen in Fig. 1 the pressure derivatives of resistivity are positive below the Curie point and negative above (see also Ref. 3). This implies a dominant role of order parameter fluctuations in the electron scattering in MnSi. Hence, any analysis of the resistivity behavior in MnSi should take into account this significant factor. We will discuss this issue elsewhere. However, it is important to emphasize here that the residual resistivity of MnSi, derived from reasonable extrapolations, decreases monotonically from 2.25 to 2.11 $\mu\Omega$ cm over all the pressure range studied on compression. This indicates that many cycles of pressure loading and unloading, cooling and warming do not introduce additional defects into the sample. The temperature-dependent resistivity of MnSi above the phase transition line shows clear signs of resistivity saturation at $T\rightarrow\infty$.[20]

Now we turn to an analysis of the temperature coefficient of resistivity $d\rho/dT$ in the vicinity of the phase transition boundary. Temperature derivatives of resistivity ρ were taken by averaging the slopes of two adjacent points of the raw experimental data. The outcome of this procedure is illustrated in Fig. 2, where also the smoothing lines are shown. As is seen from the figure at ambient pressure the curve $(d\rho/dT)(T)$ has a distinct shoulder on the high-temperature side of T_c which disappears at high pressure. The evolution of the shape of the peaks of $d\rho/dT$ with applied pressure is shown in Fig. 3. The shoulder in $(d\rho/dT)(T)$ vanishes at a pressure of around 0.35 GPa that was recognized early as a coordinate of the tricritical point.[4] The overall trend is that at

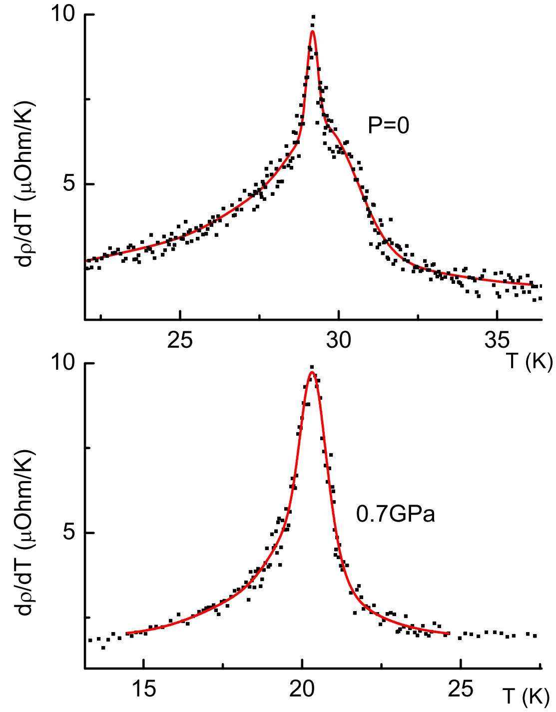

FIG. 2. (Color online) Examples of temperature derivatives of resistivity $d\rho/dT$ at ambient and elevated pressures. The square dots are the temperature derivatives of resistivity, taken by averaging the slopes of two adjacent points of the raw experimental data. The solid lines are results of smoothing procedures.

low pressure the structure in $d\rho/dT$ consists of two components: one sharp and another broad, separated only by half a degree or so. Because of lack of *a priori* knowledge of the peak forms and uncertainty with background subtraction, we could not separate these peaks in a reliable way. The obvious overlapping of the peaks makes also unreliable attempts to obtain a critical exponent, based on the behavior of

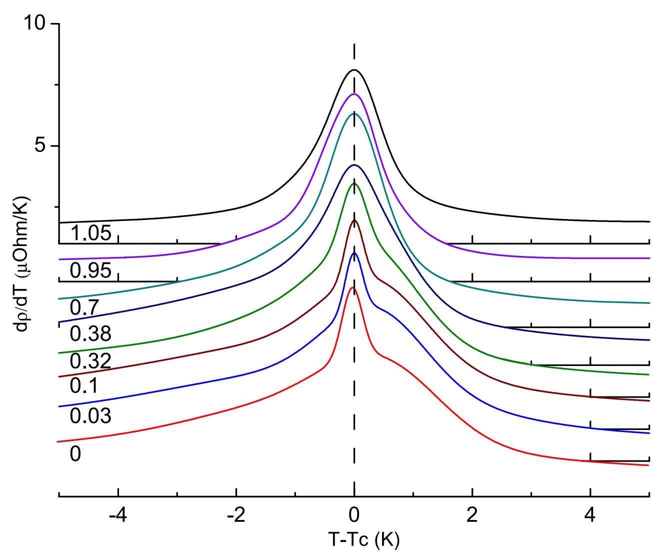

FIG. 3. (Color online) Evolution of temperature derivatives of resistivity $d\rho/dT$ with pressure. The pressures in GPa are shown at the left side of the figure.

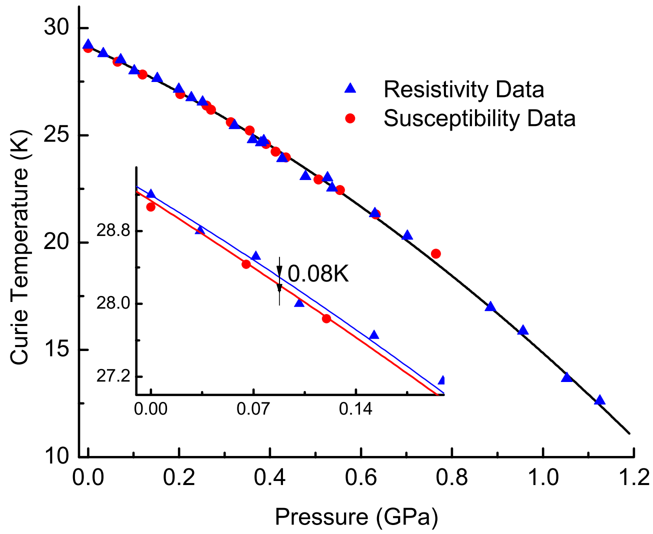

FIG. 4. (Color online) Pressure dependence of the Curie temperature of MnSi according to the current resistivity measurements and the ac susceptibility data (Ref. 4). The inset shows that the average mismatch of the two sets of the data is less than 0.1 K.

$d\rho/dT$.[10–12] Nevertheless, we have found that an approximation of $d\rho/dT$ at $T<T_c$ with the expression

$$\frac{d\rho}{dT} = a + bT + c(T_c - T)^{-m} \tag{1}$$

gives $m\approx 0.25$ in the case of the low-pressure isobars, which is a reasonable value for an exponent characterizing the critical behavior of the heat capacity near helical spin ordering.[21,22] At pressures more than 0.3–0.4 GPa, the fitting became unstable and did not lead to realistic values of the exponents.

Summarizing, we point out that the reported experimental data demonstrate complicated behavior of the temperature coefficient of resistivity of MnSi in the vicinity of its phase transition. As is seen from Fig. 3, $d\rho/dT$ evolves from a highly asymmetric, not quite resolved doublet with one rather sharp component at ambient pressure to the single, fairly symmetric peak at pressure, corresponding to the tricritical point. It was mentioned earlier that a doublet structure of related peaks was discovered in the sound absorption,[13] thermal expansion,[14] and heat capacity[15] at the phase transition in MnSi, which correlate with the current observations. Unfortunately, little is known about the origin of this structure, but what we know is that the high-temperature satellite does not reveal itself in magnetic susceptibility measurements.[3,4] The data comparison shows that the magnetic transition is associated with a sharp peak on the low-temperature side of $d\rho/dT$ (Fig. 4). Thus, the observed shoulder in $d\rho/dT$ could be connected with short-range spin order or with the spin texture.[8,9] However, it does not appear that the shoulder in $d\rho/dT$ marks any kind of conventional phase transition. Nevertheless, one cannot exclude that a topological phase transition takes place at a temperature above the magnetic transformation. In the latter case, instead of a tricritical point there would be a special kind of multicritical point in the phase diagram of MnSi. But, if the scenario with a topological phase transition is not appropriate, then the shoulder in $d\rho/dT$ disappears, being absorbed by the volume instability gap, which is opened at a tricritical point.[23]

Finally, we have observed a distinct shoulder on the high-temperature side of the peaks in the temperature dependence of resistivity at the ferromagnetic phase transition line. No doubt the indicated feature has a fluctuation origin though the nature of this fluctuation is certainly different from the fluctuations of the order parameter, normally occurring around the Curie point. Further investigations of the tricritical behavior, Fermi-liquid breakdown, and specifics of the spin order in the paramagnetic phase may provide crucial clues for the underlying physics.

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*Electronic address: sergei@hppi.troitsk.ru

[1] J. D. Thompson, Z. Fisk, and G. G. Lonzarich, Physica B **161**, 317 (1989).
[2] C. Pfleiderer, G. J. McMullan, and G. G. Lonzarich, Physica B **206-207**, 847 (1995).
[3] C. Pfleiderer, G. J. McMullan, S. R. Julian, and G. G. Lonzarich, Phys. Rev. B **55**, 8330 (1997).
[4] A. E. Petrova, V. Krasnorussky, John Sarrao, and S. M. Stishov, Phys. Rev. B **73**, 052409 (2006).
[5] N. Doiron-Leyraud, I. R. Walker, L. Taillefer, M. J. Steiner, S. R. J. R. Jullan, and G. G. Lonzarich, Nature (London) **425**, 595 (2003).
[6] C. Pfleiderer, D. Reznik, L. Pintschovius, H. v. Löhneysen, M. Garst, and A. Rosch, Nature (London) **427**, 227 (2004).
[7] Sumanta Tewari, D. Belitz, and T. R. Kirkpatrick, Phys. Rev. Lett. **96**, 047207 (2006).
[8] B. Binz, A. Vishwanath, and V. Aji, Phys. Rev. Lett. **96**, 207202 (2006).
[9] A. N. Bogdanov, U. K. Rößler, and C. Pfleiderer, Physica B **359-361**, 1162 (2005); U. K. Rößler, A. N. Bogdanov, and C. Pfleiderer, cond-mat/0603103 (unpublished).
[10] V. M. Nabutovskii and A. Z. Patashinskii, Fiz. Tverd. Tela (Leningrad) **10**, 3121 (1968).
[11] M. E. Fisher and J. S. Langer, Phys. Rev. Lett. **20**, 665 (1968).

[12] T. G. Richard and D. J. W. Geldart, Phys. Rev. Lett. **30**, 290 (1973).
[13] S. Kusaka, K. Yamamoto, T. Komatsubara, and Y. Ishikawa, Solid State Commun. **20**, 925 (1976).
[14] M. Matsunaga, Y. Ishikawa, and T. Nakajima, J. Phys. Soc. Jpn. **51**, 1153 (1982).
[15] C. Pfleiderer, J. Magn. Magn. Mater. **226-230**, 23 (2001).
[16] A. E. Petrova, V. A. Sidorov, and S. M. Stishov, Physica B **359-361**, 1463 (2005).
[17] The normal experimental procedure starts with cooling the high-pressure cell, containing a certain amount of compressed helium and the sample. After a while helium crystallizes, blocking the high-pressure tubing (Refs. 4 and 16) and making further cooling isochoric with respect to solid helium. So the cooling and subsequent warming of the sample of MnSi, which has compressibility quite different from that of helium, are neither isobaric nor isochoric.
[18] Toru Moria, *Spin Fluctuations in Itinerant Electron Magnetism* (Springer-Verlag, Berlin, 1985).
[19] Frank J. Blatt, *Physics of Electronic Conduction in Solids* (McGraw-Hill, New York, 1968).
[20] H. Wiesmann, M. Gurvitch, H. Lutz, A. Ghosh, B. Schwarz, Myron Strongin, P. B. Allen, and J. W. Halley, Phys. Rev. Lett. **38**, 782 (1977).
[21] H. T. Diep, Phys. Rev. B **39**, 397 (1989).
[22] Using the hyperscaling relation $d\nu = 2-\alpha$ and the ambient pressure value of $\nu = 0.62$ for MnSi, one finds $\alpha = 0.14$ [S. V. Grigoriev, S. V. Maleyev, A. I. Okorokov, Yu. O. Chetverikov, R. Georgii, P. Böni, D. Lamago, H. Eckerlebe, and K. Pranzas, Phys. Rev. B **72**, 134420 (2005)].
[23] At a first-order phase transition, a volume change occurs since a homogeneous state of matter becomes unstable in a certain range of volumes, which may be called the volume gap. Specific features of the behavior around a second-order phase transition could fall in the volume gap when the transition becomes first order, and therefore may not be observable.