

## High-pressure study of the magnetic phase transition in MnSi

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Measurements of ac magnetic susceptibility and dc resistivity of a high-quality single-crystal MnSi were carried out at high pressure making use of helium as a pressure medium. The form of the ac magnetic susceptibility curves at the magnetic phase transition suddenly changes upon helium solidification. This implies strong sensitivity of magnetic properties of MnSi to nonhydrostatic stresses and suggests that the early claims on the existence of a tricritical point at the phase-transition line are probably a result of misinterpretation of the experimental data. At the same time resistivity behavior at the phase transition does not show such a significant influence of helium solidification. The sharp peak at the temperature derivative of resistivity, signifying the first-order nature of the phase transition in MnSi successfully survived helium crystallization and continued the same way to the highest pressure.

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As it was found long ago, the intermetallic compound MnSi acquired a long period helical magnetic structure at  $T \approx 29$  K.<sup>1,2</sup> Itinerant nature of magnetism in MnSi was established in Ref. 3. Experiments on the influence of high pressure on the phase transition in MnSi showed that the transition temperature decreased with pressure and tended to zero at about 1.4 GPa.<sup>4</sup> This feature of MnSi promised the opportunity of observation of quantum critical behavior.

Since then, quite a number of papers have been devoted to high-pressure studies of the phase transition in MnSi (see, for instance, Refs. 5–15). Among them one should point at a paper of Pfeleiderer *et al.*<sup>5</sup> (see also Ref. 9) claimed the discovery of a tricritical point at the phase-transition line of MnSi based on the evolution of the ac magnetic susceptibility of MnSi ( $\chi_{ac}$ ) with pressure. Later on a theory was developed that declared generic nature of first-order character of phase transitions in ferromagnets at low temperatures.<sup>6</sup>

Non-Fermi liquid behavior has been observed in an extended region of pressure above the quantum critical point.<sup>11</sup> The specific magnetic structure (partial order) of the non-Fermi-liquid phase of MnSi was described in Ref. 12. Phase inhomogeneity in the region surrounding the transition line from about 10 K to the lowest temperatures was reported in Refs. 13 and 14. Incidentally, studies of the ac magnetic susceptibility of MnSi at high pressure<sup>15</sup> using fluid and solid helium as a pressure medium showed somewhat different results from the data.<sup>5</sup> In particular Petrova *et al.*<sup>15</sup> concluded that the radical change in the ac magnetic susceptibility of MnSi with pressure, observed in Ref. 5, could be influenced by nonhydrostatic stresses, developing in a frozen pressure medium. Precise lattice-constant measurements at high pressure seemingly indicate that the phase transition in MnSi is first order.<sup>16</sup>

Meanwhile studies of thermodynamic and transport properties of a high-quality single crystal of MnSi at ambient pressure suggested also a first-order nature of the corresponding magnetic phase transition<sup>17–19</sup> that again questioned the early proposed phase diagram.<sup>5–9</sup>

Having a high-quality single crystal of MnSi, which reveals sharp features of the phase transition, it was appealing to conduct studies of the phase diagram of MnSi with helium

as a pressure medium. In this Rapid Communication we report results of measurements of resistivity and ac magnetic susceptibility of MnSi at high pressure, created by compressed helium.

The samples for the current measurements as well as for an earlier study<sup>17</sup> were cut from a single crystal of MnSi, grown from melt by the Bridgman method. The resistivity was measured by a four-terminal dc method. Measurements of the ac magnetic susceptibility were carried out with a standard modulation technique at a modulation frequency of 19 Hz. Temperature was measured by a calibrated Cernox sensor imbedded in the cell body. Accuracy of the Cernox sensor in the temperature range under study is about 0.02 K. Pressure was measured by a calibrated manganin gauge while helium was still in the fluid phase. Pressure in solid helium was calculated using its known equation of state. Details of the experimental techniques are described in Refs. 15 and 20–22. Results of the experimental studies are illustrated in Figs. 1–6.

As is seen in Figs. 1 and 2, the sharp maximum of ac

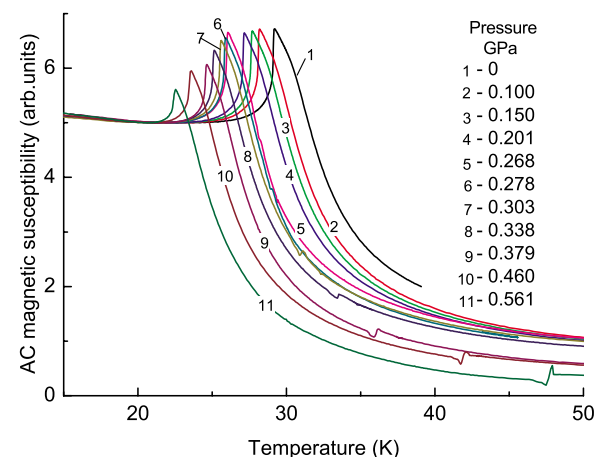


FIG. 1. (Color online) Influence of pressure on behavior of the ac magnetic susceptibility ( $\chi_{ac}$ ) at the phase transition in MnSi. Zigzag features in some of the curves are results of the helium crystallization.

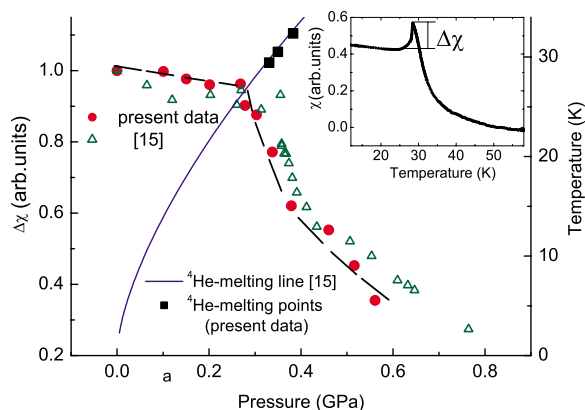


FIG. 2. (Color online) Evolution of the form of  $\chi_{ac}$  at the phase transition in MnSi. The definition of  $\Delta\chi$  is given in the inset.

magnetic susceptibility  $\chi_{ac}$  does not change much up to a pressure of about 0.3 GPa and then starts to decrease rapidly at higher pressures, as observed before.<sup>15</sup> But one could notice subtle differences between present data and the data in Ref. 15 (see Fig. 2). As it is evident from Fig. 2, a drastic change in the form of the magnetic susceptibility curve  $\chi_{ac}$  in the current data closely coincides with the helium melting point that clearly indicates the effect of nonhydrostatic stresses arising on helium solidification. A slightly different behavior of  $\chi_{ac}$  observed in Ref. 15 is most probably connected with a smaller size of the sample used in the old measurements compared to the present ones. So the nonhydrostatic stress amplitude could reach a critical value only at some distance from the melting point inside the solid helium domain. This situation led to ignoring possible nonhydrostatic effects in the experiments<sup>15</sup> with subsequent misinterpretation of the experimental data. The current measurements unambiguously show that the striking change in the magnetic susceptibility curve is a result of nonhydrostatic stresses in pressure media and has nothing to do with a change in character of the phase transition in MnSi.<sup>5,15,23</sup>

Now we turn to Fig. 3 where temperature derivatives of resistivity  $d\rho/dT$  in the vicinity of the phase transition are

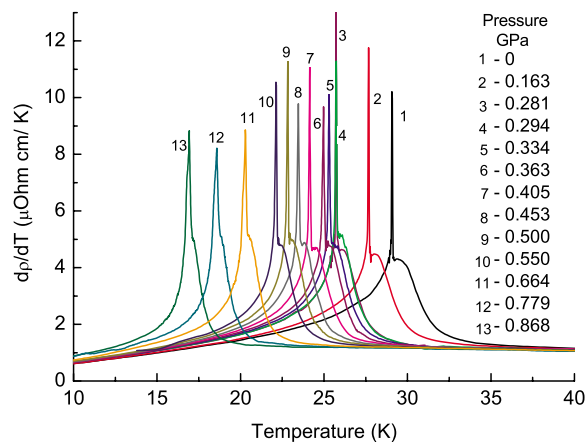


FIG. 3. (Color online) Temperature dependence of derivative of resistivity of MnSi ( $d\rho/dT$ ) at different pressures. The melting curve of helium and the phase-transition line in MnSi cross at 0.27 GPa.

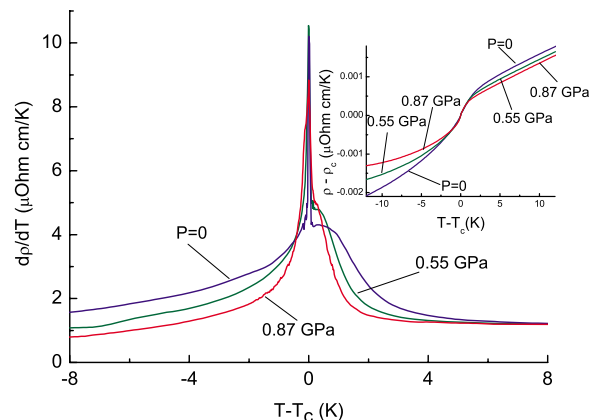


FIG. 4. (Color online) Narrowing of the global maximum of the temperature dependence of resistivity of MnSi ( $d\rho/dT$ ) at high pressures. The inset illustrates the nature of narrowing of  $d\rho/dT$ .  $T_c$  and  $\rho_c$  are the phase-transition temperature and the value of resistivity at the transition point.

depicted as a function of pressure. As one can see,  $d\rho/dT$  does not experience so dramatic a change across the helium melting line as the ac magnetic susceptibility does. However, two obvious trends are seen; both of them are better illustrated in Fig. 4. The first trend is broadening the sharp peaks of  $d\rho/dT$  that become obvious at a pressure of 0.66 GPa. As was discussed earlier,<sup>17</sup> sharp peaks of  $d\rho/dT$  originate from the first-order nature of the phase transition in MnSi. Hence, broadening of the peaks in  $d\rho/dT$  at high pressure implies smearing the phase transition by nonhydrostatic stresses. The second one is narrowing of the global anomaly in  $d\rho/dT$ , which accompanies the phase transition and reveals itself as a satellite rounded peak on the high-temperature side of the phase transition. Note that the  $d\rho/dT$  anomaly scales perfectly with the corresponding anomalies in heat capacity and thermal-expansion coefficient data of MnSi.<sup>17,19</sup> Figure 5 shows an evolution of the width of maxima in  $d\rho/dT$  on pressure increasing. Surprisingly, in contrast with the case of

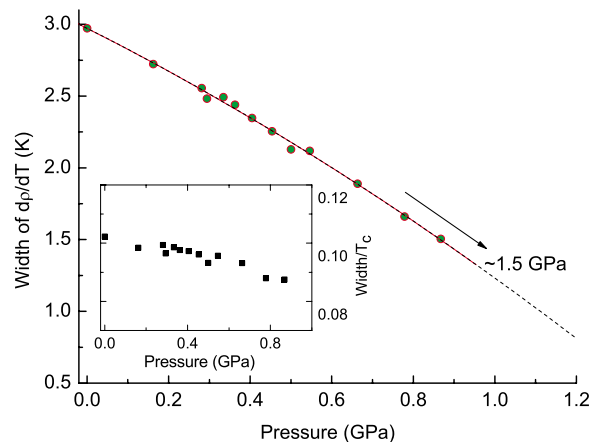


FIG. 5. (Color online) Pressure dependence of the width of the global maximum in  $d\rho/dT$ . The inset shows small variation in the ratio width/temperature with pressure. The width is taken at the level of 3  $\mu\text{Ohm cm/K}$  (see Fig. 3). Note that the melting curve of helium and the phase-transition line are crossed at about 0.27 GPa.

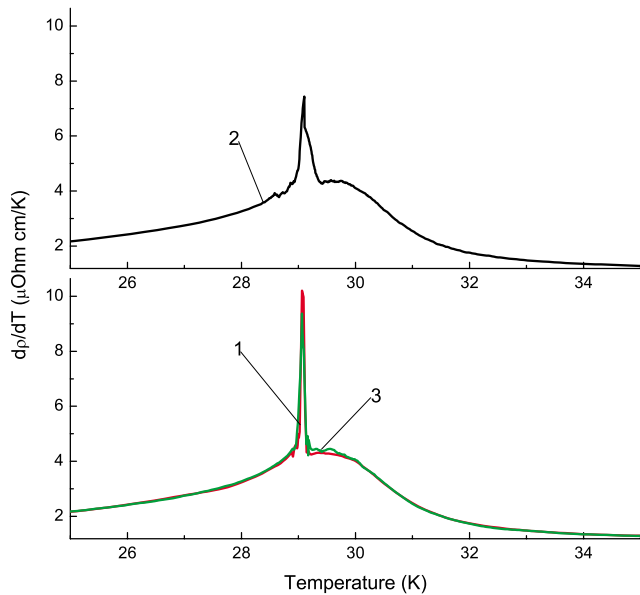


FIG. 6. (Color online) Influence of fast pressure releasing on the peak of  $dp/dT$  at the phase transition in MnSi. 1—ambient pressure, 2—ambient pressure after loading the sample to 1.2 GPa, and 3—after holding the sample experienced 1.2 GPa for a week at ambient pressure.

magnetic susceptibility, no trace of the helium crystallization is seen on the corresponding curve. As it follows from Figs. 3 and 4 narrowing of the anomaly signifies its general reduction despite the fact that its amplitude slightly increases.<sup>24</sup> More specifically this implies decrease of the general abundance of spin fluctuations along the transition line since electron scattering on spin fluctuations provides a major contribution to the resistivity of MnSi around the phase transition. Simple extrapolation of the curve in Fig. 5 to the zero width leads to the pressure value about 1.5 GPa, which almost exactly corresponds to the phase-transition pressure at  $T=0$ , as should be expected.

Another effect observed in the current study of resistivity of MnSi is defect generation upon pressure releasing, as is illustrated in Fig. 6. It is seen that, after decreasing pressure from  $\sim 1.2$  GPa to zero in a time span of less than 1 h, the peak of  $dp/dT$  appears to be highly distorted. Remarkably, after holding the sample for a week at ambient pressure and

room temperature, the form of the peak completely recovered its initial shape. All that did not happen when the sample was loaded by 0.7 GPa of helium pressure. This observation tells us that at some pressure, obviously more than 0.7 GPa, MnSi experiences some sort of irreversible change on a short time scale that facilitates generation of defects on pressure releasing. Despite all these complications and even ignoring the data obtained above 0.7 GPa, we are still able to derive certain conclusions with regard to the phase transition in MnSi at high pressure.

First, nonhydrostatic stresses arising in solid helium strongly influence the form of ac magnetic susceptibility of MnSi and the latter cannot serve as an indicator of the type of phase transition. So, claims of the existence of a tricritical point on the phase-transition line in MnSi seem to be a result of misinterpretation of the experimental data.<sup>5,10,15</sup> Second, the electrical resistivity of MnSi appears to be almost insensitive to the nonhydrostatic stresses and the persistent sharp peak of  $dp/dT$  experiences only slight broadening with pressure. Third, the existence of  $dp/dT$  peaks at the highest pressures provides evidence that the magnetic phase transition in MnSi does not change its nature in the whole pressure range studied and most probably will stay the same way with further increase in pressure.

Finally, the different sensitivity of ac magnetic susceptibility and electrical resistivity to nonhydrostatic environment probably indicates the dual role of the spin fluctuations involved in the corresponding physics. But detailed analysis of the situation should wait until there is a proper understanding of the nature of the satellite rounded maxima in  $dp/dT$  as well as the heat capacity and the thermal-expansion coefficient at the phase transition in MnSi.<sup>25</sup>

*Note added in proof.* Recent study of the iron arsenide compound  $\text{CaFe}_2\text{As}_2$  at high pressure created by frozen liquid and helium<sup>28</sup> clearly demonstrates the influence of nonhydrostatic conditions on the observation of physical effects.

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  - <sup>23</sup>Holding the cell at 35 K and at 0.5 GPa pressure of solid helium for more than 12 h in an attempt to reduce nonhydrostatic stresses did not affect the magnetic susceptibility of the sample.
  - <sup>24</sup>Eventually, the maximum in the  $d\rho/dT$  and the  $d\rho/dT$  itself disappears in the low temperature limit by the general law.
  - <sup>25</sup>It is tempting to connect these rounded maxima with a diffuse ring, observed in neutron-scattering experiments in the paramagnetic phase of MnSi, indicating the appearance a quasilong-range spin order at temperature slightly above  $T_c$  (Refs. [26](#) and [27](#)).
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