

Reply to “Comment on ‘Nature of the high-pressure tricritical point in MnSi’ ”

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In this Comment we give a detailed reply to the criticisms made by Stishov in the accompanying comment to our paper [Otero-Leal *et al.*, Phys. Rev. B **79**, 060401 (2009)]. We demonstrate that the magnetic phase transition occurring at pressures larger than 3.5 kbar in MnSi is of the first order and that the classical Landau’s analysis of continuous phase transitions captures these effects.

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The location of a tricritical point in the (P, T) phase diagram of MnSi and the nature of the phase transition at both sides of this point are under discussion. While the transition has been originally located at $p_{tc}=12$ kbar and $T_{tc}=12$ K, with the first-order transition at $p > p_{tc}$, Stishov *et al.*¹ suggested that it should be located at $p_{tc}=3.5$ kbar and $T_{tc}=25$ K with the transition being second order for $p > p_{tc}$. This is an important difference, as it could change the common understanding of the phase diagram of this prototypical weak itinerant ferromagnet. Moreover, Belitz *et al.*^{2,3} derived a mean-field expression for the free-energy density of weak itinerant magnets and demonstrated that at low temperatures, the coupling between the particle-hole excitations and the fluctuations of the magnetic order parameter lead to a change in the order of the magnetic phase transition and hence to a tricritical point in the P, T phase diagram. The contradictory results reported in the literature about the nature of the high-pressure phase transition in MnSi also prevent the experimental evaluation of this model.

However, most of the conclusions about the order of the phase transition are based on indirect qualitative proofs (disappearance of the resistivity or susceptibility peak, etc.), which could be dependent on the sample, field used, etc. In order to solve this controversy Otero-Leal *et al.*⁴ performed an study in which they determined the order of the magnetic phase transition from magnetic measurements, through the analysis of the experimental $\chi(H)$ in the spirit of Landau’s classic treatment of second-order magnetic phase transitions.⁵ This study demonstrated that the tricritical point in MnSi is located at $p_{tc}=3.5(3)$ kbar and $T_{tc}=25$ K, and more important, the transition changes from second-to-first order as pressure increases, in agreement with the scenario proposed by Belitz *et al.*^{2,3}

However, in the accompanied comment to the paper by Otero-Leal *et al.*,⁴ Stishov finds our conclusions and analysis erroneous. The comment is focused in two issues: (1) The effect of nonhydrostaticity over the evolution of the magnetic susceptibility. (2) The validity of Eq. (1) in our paper to analyze the change in the order of the magnetic phase transition of MnSi. Actually both points are related in this case, as Stishov proposes that the change reported in the nature of the magnetic phase transition is an artifact produced by nonhydrostatic conditions during our experiments.

As it is written in the experimental part of our paper we used Daphne 7373 paraffin industrial oil which does not un-

dergo any change at this particular pressure or temperature. Although once solidified the oil will not transmit pressure in ideal hydrostatic conditions, the point here is that there is no change in the state of the transmitter and hence on the hydrostatic conditions at this particular pressure/temperature point. Note that we have avoided the use of He as a pressure transmitter in our study because it solidifies precisely at the tricritical point of MnSi, introducing an important source of nonhydrostaticity.

On the other hand, the change in the curvature of the H/M vs M^2 curves in Fig. 2 of our paper is very smooth across the tricritical point, not consistent with a sudden change in the physical state of the paraffin oil used as pressure medium. These effects are reversible and reproducible from sample to sample. We have also used Daphne 7373 oil to study the critical behavior of different systems under pressure (for example, CoS₂, etc), obtaining completely different locations of the tricritical point.⁶ Moreover, it is important to note that Pfeleiderer *et al.*⁷ independently observed the presence of a tricritical point in the phase diagram of MnSi using a different oil (n-pentane) as the transmitting medium.

So, this should be enough to demonstrate that the changes reported in our paper at this particular (P, T) cannot be due to a change in hydrostatic conditions of the pressure medium at this particular pressure/temperature. Regarding the second point of his comment, that is, the use of Eq. (1) in our paper to analyze the phase transition in MnSi.

The main motivation for our work was the existence of two papers by Belitz and co-workers^{2,3} in which they derived a mean-field expression for the free-energy density of weak itinerant magnets. One of the results of their model is that at low temperatures, the coupling between the particle-hole excitations and the fluctuations of the magnetic order parameter lead to a change in the order of the magnetic phase transition and hence to a tricritical point in the P, T phase diagram (at high pressure, when T_c is sufficiently suppressed). In their Landau-type theory this is manifested as a change in the sign of the quartic term of the free energy vs order-parameter expansion. This is exactly the basis of the analysis we have performed in our paper.

But the point of Stishov in his comment to our paper is that because of the helical order of MnSi, this model is not applicable (the magnetization is not the correct order parameter in this case). However, in a more recent paper Gehring⁸ uses a Landau theory to map out the phase diagram of MnSi.

He shows that a first-order phase transition is always predicted before the quantum critical point, in agreement with our results and with previous theoretical predictions of Belitz and co-workers. But more important, Gehring shows that this will occur for any type of magnetic order, not just ferromag-

netic, and without including fluctuations. So, the use of a Landau-type model to follow the evolution of the nature of the phase transition in MnSi is fully justified and the existence of a tricritical point in the phase diagram of MnSi well demonstrated.

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