Comparative study of helimagnets MnSi and Cu$_2$OSeO$_3$ at high pressures

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The heat capacity of helical magnets Cu$_2$OSeO$_3$ and MnSi has been investigated at high pressures by the ac-calorimetric technique. Despite the differing nature of their magnetic moments, Cu$_2$OSeO$_3$ and MnSi demonstrate a surprising similarity in behavior of their magnetic and thermodynamic properties at the phase transition. Two characteristic features of the heat capacity at the phase transitions of both substances (peak and shoulder) behave also in a similar way at high pressures if analyzed as a function of temperature. This probably implies that the longitudinal spin fluctuations typical of weak itinerant magnets like MnSi contribute little to the phase transition. The shoulders of the heat capacity curves shrink with decreasing temperature suggesting that they arise from classical fluctuations. In case of MnSi the sharp peak and shoulder at the heat capacity disappear simultaneously probably signifying the existence of a tricritical point and confirming the fluctuation nature of the first order phase transition in MnSi as well as in Cu$_2$OSeO$_3$.

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I. INTRODUCTION

Manganese silicide (MnSi), a model itinerant helimagnet, crystallizes in a B20 structure, whose non-centrosymmetric space group P2$_1$$^3$ allows a helical (chiral) magnetic structure. The phase transition in MnSi from helical to paramagnetic states at $\approx$30 reveals some remarkable features such as sharp peaks and shoulders on the high temperature side of the peaks in the heat capacity, thermal expansion, temperature dependence of resistivity and sound absorption$^{1,2}$. As is recognized now the sharp peaks in the above properties indicate a first order nature of the phase transition whereas the origin of the shoulders is still not quite clear$^{1,5}$. There is some evidence that the shoulders arise from intense helical (chiral) fluctuations in the vicinity of the phase transition in MnSi$^{3,4}$. These fluctuations must be responsible for the first order phase transition in MnSi, symmetry of which principally allows a second order one$^{11,12}$. The simultaneous disappearance of the shoulder and first order features at the phase transition in MnSi on decreasing temperature supports this conclusion$^{13}$.

Insulator Cu$_2$OSeO$_3$ crystallizes in a complicated structure, which belongs to the same space group P2$_1$$^3$ common to MnSi, and hence permits piezoelectricity$^{13}$. On cooling below 60 K Cu$_2$OSeO$_3$ becomes magnetically ordered and demonstrates an enhanced magneto dielectric effect. The latter implies existence of the magneto electric coupling in the system. Careful structural studies of Cu$_2$OSeO$_3$ show no measurable structural distortion occurring down 10 K. This means that there is no a spontaneous lattice distortion involved in the magneto electric coupling mechanism. So a pure electronic coupling should be responsible for the observed magneto dielectric response$^{13}$. These observations predict an incommensurate magnetic structure$^{13}$. Finally, a helical spin structure in Cu$_2$OSeO$_3$ was found in a small angle neutron scattering experiment$^{15}$. But despite a similar spin structure, major differences between metallic MnSi and insulating Cu$_2$OSeO$_3$ lies in their magnetic moments. The moments are itinerant in MnSi and local in Cu$_2$OSeO$_3$. Nevertheless the behavior of the heat capacity and magnetic susceptibility at the magnetic phase transitions in MnSi and Cu$_2$OSeO$_3$ appears to be very similar. Indeed the heat capacity of Cu$_2$OSeO$_3$ shows a peak and shoulder on the high temperature side of the peak at the phase transition like the heat capacity of MnSi.

Actually the difference in nature of the magnetic moments between the two substances noted above reveals itself in a pressure dependence of the phase transition temperature $T_c$. $T_c$ of MnSi decreases with pressure and tends to zero, whereas $T_c$ of Cu$_2$OSeO$_3$ increases with pressure at least up to 2 GPa$^{17}$. It would be of great interest to track an evolution of the features of the phase transitions in both materials in the hope of shedding more light on its nature and the origin of the strongly fluctuating region (shoulder). The evolution of resistivity in the vicinity of the phase transition in MnSi with pressure was analyzed in$^{41}$.

Here we report results of a high pressure study of the heat capacity of MnSi and Cu$_2$OSeO$_3$ and the ac-magnetic susceptibility of Cu$_2$OSeO$_3$. The data obtained show different pressure but similar temperature dependences of specific features of the phase transition, therefore confirming the classical character of the strongly fluctuating region and the fluctuation nature of the first order transition in MnSi at ambient and moderate pressures.

II. EXPERIMENTAL

Single crystals of Cu$_2$OSeO$_3$ of size 0.5–1.0 mm were grown by a gas transport technique in a 610 – 550°C temperature gradient using a 2:1 CuO/SeO$_2$ mixture and CuCl$_2$$\cdot$2H$_2$O as a transport agent. High pressures were
The two substances under study belong to different classes of solids. Cu$_2$OSeO$_3$ is a covalent insulator with local magnetic moments, whereas metallic MnSi is an itinerant magnet. Normally the exchange interaction, giving rise to magnetic order in a system with local magnet moments depends on inter-particle distances in such a way that increases a phase transition temperature with pressure. This is what happens with $T_c$ of Cu$_2$OSeO$_3$ with applied pressure (see Fig. 2b). In the case of itinerant magnets an energy gain occurring at magnetic ordering arises as a result of competition between the exchange interaction and electron kinetic energy. Their changes on compression lead to a decrease of the transition temperature in itinerant magnets with pressure (see Fig. 2b). Despite the mentioned difference both substances demonstrate a surprising similarity in behavior of magnetic and thermodynamic properties at the phase transition (Figs. 1a, b). Probably, this implies that the longitudinal spin fluctuations typical of the weak itinerant magnets do not contribute much to the discussed properties.

Remarkably the similarity between Cu$_2$OSeO$_3$ and MnSi found at ambient pressure can be seen at high pressures as well, if one uses temperature as a variable. Indeed, analyzing Figs. 4 and 5 which depict the behavior of the anomalous part of heat capacity of Cu$_2$OSeO$_3$ and MnSi with pressure and temperature. The length of the sample of Cu$_2$OSeO$_3$ decreases linearly under pressure up to 4.5 GPa. The bulk modulus calculated from the change of the sample length is equal to 197 ± 2 GPa.
The heat capacity of helical magnets Cu$_2$OSeO$_3$ and MnSi has been studied at high pressures by the ac-calorimetric technique. The magnetic ac-susceptibility was measured in the vicinity of the magnetic phase transition in Cu$_2$OSeO$_3$. The helical phase transition temperature $T_c$ increases with pressure in case of Cu$_2$OSeO$_3$, whereas $T_c$ of MnSi drops on compression in accordance

along the phase transition line towards lower temperatures, irrespective of the different pressure dependence. Narrowing the splitting is a consequence of shrinking the heat capacity anomaly (shoulder) along with the transition temperature reduction. All this helps to identify the shoulder as a product of classical fluctuations. At the same time, in case of MnSi the sharp peaks, which classify the transition as first order cease to exist at low temperatures almost simultaneously with the disappearance of shoulder. Then with further decrease of temperature the heat capacity peaks are progressively reduced in size and width (Figs. 5, 6). This behavior leads us to claim the existence of a tricritical point in the phase diagram of MnSi, as is shown in Fig. 7b, in agreement with conclusions of Ref. 13.

V. CONCLUSION

FIG. 2: (Color online) Pressure dependence of the magnetic phase transition temperature in Cu$_2$OSeO$_3$ (a) 1- ac-susceptibility, 2- ac-calorimetry, sample 1, 3- ac-calorimetry, sample 2; 4-Ref. 17 and MnSi (b) 1-Ref. 21, 2-Ref. 22.

FIG. 3: (Color online) Magnetic susceptibility $\chi$ of Cu$_2$OSeO$_3$ at different pressures.

FIG. 4: (Color online) Anomalous part of the heat capacity of Cu$_2$OSeO$_3$ at different pressures.

FIG. 5: (Color online) Anomalous part of the heat capacity of MnSi at different pressures.
FIG. 6: (Color online) Temperature derivatives of the resistivity $d\rho/dT$ at the phase transition in MnSi at different pressures after Ref. 13.

FIG. 7: (Color online) Temperature difference $\Delta T$ between the peaks and the shoulder maxima as a function of pressure.

FIG. 8: Phase diagrams of Cu$_2$OSeO$_3$ and MnSi.

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