UNCONVENTIONAL ANTIFERROMAGNETISM OF Mn_3Si AND CuMnSb*

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We report an investigation of the antiferromagnetic Heusler metal Mn_3Si and semi-Heusler metal CuMnSb. The Néel-temperatures $T_N \approx 23$K for Mn_3Si and $T_N \approx 50$K for CuMnSb as seen by a pronounced anomaly in the specific heat are not affected by a magnetic field up to 14 T. The magnetisation is unsaturated, but does not show any signs of metamagnetic transitions in the ordered state. The resistivity drops at the onset of antiferromagnetism, but is not affected by magnetic field apart from the residual value for $T \to 0$, which increases slightly. This invariance of the antiferromagnetic order to high magnetic field is incompatible with present day models for magnetism in metals, and suggests an unconventional form of antiferromagnetic order.

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A longstanding controversy in the theory of magnetic metals is about the question, how to generalize the Fermi liquid model of paramagnetic metals to describe magnetic order. Weakly magnetic transition metal compounds with small ordered moments and low transition temperatures are well described in terms of a Fermi liquid model with exchange split Fermi surface [1, 2]. In contrast, it is still an open issue if magnetic metals with large ordered moments and small ordering temperature that are more akin to local moment insulators, may also be described on the basis of Fermi liquid theory.

Here we review the properties of two transition metal compounds, Mn_3Si and CuMnSb, that exhibit antiferromagnetic order of low ordering temperatures and large ordered moments in a metallic state [3, 4]. Both materials belong to the general class of Heusler alloys that usually develop ferromagnetism at temperatures as high as 1000 K. In our studies we find that the specific heat, high field susceptibility and electrical resistivity are not affected

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by high magnetic field up to 14 T. This is not expected in the framework of an antiferromagnetic Fermi liquid and suggests a novel class of antiferromagnetic metals.

Mn_3Si crystallizes in the Heusler structure DO_3 as $Mn_IMn_{II,2}Si$. Its crystal structure may be described in terms of four f.c.c. sublattices located at (0, 0, 0) for Mn_I, (1/4, 1/4, 1/4) and (3/4, 3/4, 3/4) for the Mn_{II} and (1/2, 1/2, 1/2) for Si. Elastic neutron scattering shows that incommensurate antiferromagnetic order develops below $T_N \approx 25$ K along $\boldsymbol{Q} = 0.425\ \boldsymbol{a}_{111}$, where (111) denotes the reciprocal lattice vector [5]. The ordered moments are $\mu_I = 1.7\mu_B$ and $\mu_{II} = 0.19\mu_B$, respectively. The specific heat $C(T)$ is dominated by a pronounced anomaly at T_N (Fig. 1), where the zero-field behaviour is consistent with that reported before and $\gamma = C/T = 69\,\mathrm{mJ/mol\,K^2}$ for $T \to 0$ [6]. The rounding of the anomaly in $C(T)$ and the slightly reduced $T_N = 23$ K may be due to the polycrystalline nature of the samples studied here. Up to a field of 14 T we do not observe any variation of the form of $C(T)$ within the resolution of the experiment, apart from a slight increase of $C(T)$ above T_N. Lattice contributions to $C(T)$ are negligibly small up to 30 K as estimated from the Debye temperature $\Theta_D = 454$ K taken from thermal expansion [7].

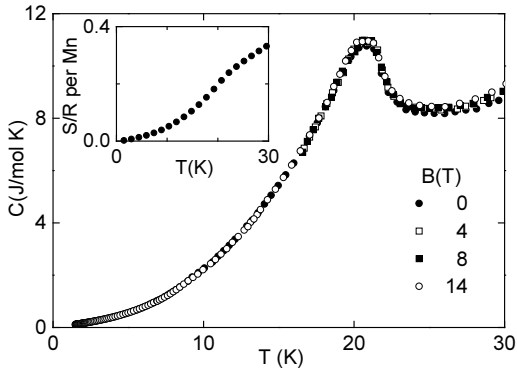

Fig. 1. Specific heat C as a function of temperature T of Mn_3Si. The anomaly corresponds with the ordering temperature at $T_N \approx 23$K. The rounding may be attributed to the polycrystalline nature of the samples studied here. T_N is not affected by high magnetic field up to 14 T, apart from a slight increase of 3% above T_N.

A key aspect of the nature of the normal metallic state above T_N and the magnetic order is the entropy $S(T)$ (inset of Fig. 1) as computed from the experimental data of $C(T)$. Near T_N the entropy reaches $R\ln 2$ per Mn_3Si, *i.e.*, it is an order of magnitude larger than observed experimentally for weakly ferromagnetic transition metal compounds [8]. In the theory

of weakly magnetic metals, the entropy is related to Lorentzian spectral distributions of the spin fluctuations. The high entropy thus fundamentally questions, if the metallic state is consistent with a spin fluctuation model based on Fermi liquid theory.

The magnetisation M as function of magnetic field B of polycrystalline samples reaches only a tiny fraction of the ordered moment up to 12 T at all T. The magnetic response is linear in B and characteristic of a simple Pauli paramagnetic state over the entire B and T range investigated [3]. The apparent absence of metamagnetic transitions underscores the lack of magnetic field dependence. This is supported further by the resistivity $\rho(T)$, which exhibits an intimate interplay of metallic state and magnetism in terms of a broad shoulder near 50 K [3,6], which however is not sensitive to magnetic field up to 12 T either, even in the immediate vicinity of T_N.

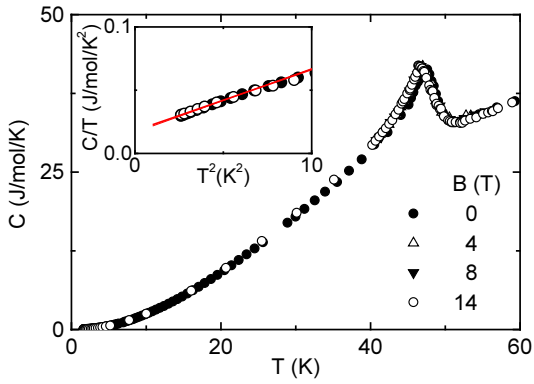

Fig. 2. Specific heat C as a function of temperature T of CuMnSb. The anomaly corresponds with the antiferromagnetic ordering temperature $T_N \approx 50$K. In small fields the transition becomes slightly sharper, probably due to the polycrystalline nature of the samples. At high magnetic field T_N remains unchanged.

The semi-Heusler compound CuMnSb crystallizes in the $C1_b$ cubic structure which differs from the DO_3 structure of Mn_3Si in that the f.c.c. sublattice at (1/4 1/4 1/4) is not occupied. CuMnSb develops antiferromagnetic order below $T_N \approx 50$K with an ordered moment of $4.0\mu_B$/f.u. [9]. The specific heat as function of temperature of polycrystalline samples (Fig. 2) exhibits a pronounced anomaly at the antiferromagnetic ordering temperature $T_N \approx 50$ K. Up to 14 T no shift of T_N is observed, apart from an initial small narrowing. The low temperature behavior may be described as $C/T = \gamma + \beta T^2$ as illustrated in the inset of figure 2, where $\gamma \approx 17$mJ/molK^2. The value of $\beta = 5.1$mJ/molK^4 at $B = 0$ and 5.4 mJ/molK^4 at $B = 14$ T exceeds the Debye lattice contribution inferred from the thermal expansion [10], $\beta_D \approx 6.5 \times 10^{-2}$ mJ/molK^4, by two orders of magnitude.

The magnetisation of CuMnSb is highly unsaturated and increases linearly as function of magnetic field up to 12 T, *i.e.*, it does not exhibit any hints of metamagnetic transitions expected of an antiferromagnet. The magnetic moment at 5 K and 12 T of $\sim 0.25\mu_B$/f.u. is an order of magnitude larger than for Mn_3Si, but remains significantly smaller than the ordered moment of $4.0\mu_B$/f.u. and the fluctuating Curie–Weiss moment derived of the high temperature susceptibility $\mu_{\rm eff} \approx 7.2\mu_B$/f.u. This underscores the unusual absence of magnetic field dependence of the antiferromagnetism and is not expected of an antiferromagnetic Fermi liquid. The electrical resistivity at $B = 0$ and 12 T decreases from room temperature to 1.5 K in agreement with [11]. At high temperatures $\rho(T)$ is only weakly temperature-dependent, but displays a pronounced drop below T_N. At a field of 12 T $\rho(T)$ is unchanged, but the high residual resistivity $\rho_0 \approx 50\mu\Omega$cm is slightly increased.

In conclusion the Heusler alloys Mn_3Si and CuMnSb develop antiferromagnetic order in a metallic environment, which despite of its low ordering temperatures is not affected by high magnetic fields. This invariance is not consistent with the properties of an antiferromagnetic Fermi liquid and may be due to a competing spin and charge density wave and/or strongly asymmetric density of states of the majority and minority charge carriers.

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