Are Mn_3Si and CuMnSb Antiferromagnetic Half-Metals?

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Abstract

The Heusler alloy Mn_3Si and the semi-Heusler alloy CuMnSb order antiferromagnetically at low temperatures. The complete absence of magnetic field dependence of the antiferromagnetic order in these compounds, inferred from the resistivity, magnetisation and specific heat are, however, incompatible with the standard model of itinerant magnetism. Here I discuss the possibility that this unusual stability may be related to a strongly reduced density of states of the minority charge carriers usually considered in the context of half-metallic ferromagnetism only.

Key words: half-metallic magnetism, itinerant antiferromagnetism, spin density wave

Strongly spin polarized band-ferromagnets are of great scientific interest for their potential use in spin-sensitive electronic devices. Candidate materials exhibiting the extreme limit of a gap in the density of states of the minority charge carriers, also referred to as half metallic *ferro*magnets (HM-FM) [1], belong mostly to the spinels, Heusler and semi-Heusler alloys such as Fe_3O_4, Co_2MnSi and NiMnSb, respectively [2]. Materials with fully spin polarized conduction electrons, but vanishing external net magnetic moment, i.e. half-metallic *antiferro*magnets, have in contrast only been considered a theoretical possibility [3]. Half-metallic antiferromagnetism is predicted to arise in the form of perfect *ferri*magnetism, i.e., sublattices of differing atomic species order antiferromagnetically without the doubling of the magnetic unit cell in conventional antiferromagnets. A compound for which half-metallic antiferromagnetism is predicted is CrMnSb, which however does not crystallize with the required $C1_b$ semi-Heusler space group [3,4].

In this paper I present plausibility arguments to show that the antiferromagnetism of the Heusler alloy Mn_3Si and the semi-Heusler alloy CuMnSb share many features expected of half-metallic antiferromagnets. Most importantly, a lack of magnetic field dependence may be readily explained, because the conduction electrons for a HM-AFM are fully spin-polarized, and spin flip excitations between the minority and majority Fermi surface are absent.

The Heusler compound Mn_3Si, space group $L2_1$, develops incommensurate spin density wave order (SDW) below $T_N = 23\,K$ [5]. The SDW in Mn_3Si may be considered a weak analogon to the order observed in Cr. Further, there are two different Mn sites ($Mn_I Mn_{II,2}Si$), for which the ordered moments are $\mu_{MnI} = 1.7\mu_B$ and $\mu_{MnII} = 0.19\mu_B$, respectively. The semi-Heusler alloy CuMnSb, space group $C1_b$, on the other hand, develops type 2 (f.c.c.) antiferromagnetic order below $T_N = 54\,K$ with $Q \parallel < 111 >$ and large ordered moments $\mu_s = 4\mu_B/Mn$ [6]. As described in detail elsewhere [7] the zero field magnetic properties of both compounds are typical of itinerant antiferromagnets. This is readily evident, for instance, in the strong decrease of the metallic resistivity below T_N.

We have recently reported an experimental study of polycrystalline samples of Mn_3Si and CuMnSb [7]. As our main result we find that despite the low Neel temperatures the antiferromagnetic state is not affected whatsoever by high magnetic field up to 14 T. This is featured by the absence of a magnetic field dependence of T_N, the specific heat and the resistivity, and an ab-

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sence of metamagnetic transitions, i.e., the stabelisation of only a very small uniform magnetisation ($M \propto B$) up to 12 T at all temperatures. To our knowledge an equivalent stability in high magnetic field has not been identified before, but may have been overlooked, for instance, in pure Cr [8] for which T_N is high.

It has been noticed in numerous theoretical studies, that the local density approximation quantitatively accounts for the electronic structure of Heusler and semi-Heusler alloys, namely the ordered magnetic moments [9]. As part of these studies it was observed that Heusler and especially semi-Heusler compounds containing Mn show a strong trend to support a fully spin polarized ferromagnetic state. Spin polarized band structure calculations for Mn_3Si, in which contributions from the two Mn sites are distinguished [10], indeed show a half-metallic state for the Mn_I site and only a very small minority d.o.s. for the Mn_{II} site.

Similar arguments may be presented for CuMnSb. Even though the band structure of CuMnSb has not been reported, a large number of AMnSb isostructural compounds have been studied, where A is a transition metal [1,2,9]. As key result it was found that for the $C1_b$ space group the point symmetry of the Mn site quite generally implies a bonding of the Mn p- and d-orbitals with the Sb p states, that causes a shift of the bands away from the Fermi level and results in a halfmetallic state.

A further key feature of half-metallic ferromagnets is that the conduction bands contain an integer number of electrons and that therefore the ordered magnetic moment is an integer number of μ_B. Mn_3Si clearly does not exhbit integer values of the ordered moments. Interestingly, though, the sum of the sublattice moments ($\mu_{\rm ord} \approx 2.08\mu_B$/f.u.) would qualify as an integer ordered moment for the case of a half-metallic ferromagnet. This may hint on subtle fluctuations in a half-metallic antiferromagnetic state for Mn_3Si. The ordered moment of CuMnSb of $4.0\mu_B$/f.u., on the other hand, is of integer value as expected.

In contrast with the predictions of half-metallic anitferromagnetism, Mn_3Si and CuMnSb exhibit the conventional doubling of the magnetic unit cell [5,6]. However, rigourous theoretical arguments [11] show that electronic systems far from half-filling are highly susceptible to a coupled spin and charge density wave instability. Since the case of a half-metallic ferromagnet represents the strongest deviation from half-filling, the antiferromagnetism in Mn_3Si and CuMnSb might hence be the result of a density wave instability and is as such not in contradiction with HM-AFM.

Further points of concern are: (i) the finite albeit weak high field susceptibility, which may suggest particle hole excitations, and (ii) the low ordering temperature, which may suggest that the antiferromagnetic spin splitting vanishes at T_N. Yet, HM-AFM are characterised by a heavily asymmetric density of states of the majority and minority charge carriers [12]. Thus for a HM-AFM it is possible in principle to achieve a large high field susceptibility driven by a shift of the Fermi level only. Secondly, strong spin fluctuations well above T_N in Mn_3Si clearly show that the spin splitting is already well developed on short time scales at high temperatures. For Mn_3Si and CuMnSb it may suggest that the ordering process is driven by the temperature dependence of the Fermi level as well and possibly further mechanisms controling long range coherence of the electronic many body wave function, but does not contradict HM-AFM as such.

It is evident that temporal and spatial fluctuations of the spin polarization in Mn_3Si and CuMnSb yield the key to an understanding of the stability of the antiferromagnetism in high magnetic field. Measurements of the spin polarisation using point contact spectroscopy and spin sensitive photoemission are therefore now in preparation.

Acknowledgements

I would like to thank J. Bœuf and A. Rosch for many stimulating discussions and H. v. Löhneysen for complete freedom to initiate and perform a research program on the magnetic properties of Heusler compounds while being a member of his group.

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