

MAGNETIC SUSCEPTIBILITIES OF Mn_5Si_3 AND $MnSi$

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Magnetic properties in relation to electronic states and bond types are especially important for the transition metals, but only very limited information is available, especially for the silicides, where the data are isolated and conflicting. For instance, it has been claimed that $MnSi_2$ is diamagnetic but Mn_2Si and $MnSi$ are paramagnetic [1] and also that all manganese silicides are ferromagnetic.

Here we report magnetic susceptibilities for stoichiometric Mn_5Si_3 and $MnSi$.

The initial materials were 99.98% Mn (twice distilled under vacuum) and chips of silicon single crystals (over 99.997% Si). The products were homogenized and then annealed under vacuum at 900°C for 20 hr. The phase composition was checked from the electrical parameters and also by metallography. The susceptibility was measured to $\pm 5\%$ or better by Faraday's method over the range 20 to 900°C with an evacuated apparatus in fields of about 5000 G.

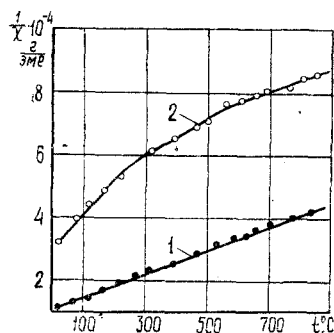


Fig. 1. Magnetic susceptibility of (1) Mn_5Si_3 and (2) $MnSi$ as a function of temperature.

Figure 1 gives the results, which show that both compounds are paramagnetic, the susceptibilities at 20°C (in 10^{-6} emu/g) being respectively 90 and 30. Mn_5Si_3 follows the Curie-Weiss law closely, the effective magnetic moment of the Mn atom being $3.9 \mu_B$ over the range 20-850°C (the Si atom is assumed not to contribute to this), with $\Theta = 0^\circ K$, which differs somewhat from the result of [3]. The monosilicide deviates from that law, and there are three distinct ranges within this temperature interval (20-250, 250-550, and 550-900°C), in which μ_{eff} is respectively 2.5, 3.3, and $4.1 \mu_B$. The result for 20-250°C agrees closely with that of [1]; the temperature effect for $MnSi$ resembles that for $CrSi$ [4] and is due to change in the electronic state of the Mn atom. The following scheme [5] differs from that in [6]: The ground state of Mn at room temperature is such that five of the seven outer electrons form bonds. One forms the bond to the nearest Si atom ($r = 2.30 \text{ \AA}$), three to the other six Si (three at 2.39 \AA , bond of order $2/3$; three at 2.55 \AA , bond of order $1/3$), and one to the six adjacent Mn atoms ($r = 2.79 \text{ \AA}$, bond

order $1/6$). All four outer electrons of the Si are involved ($1 + 3 \times (2/3) + 3 \times (1/3) = 4$). This leaves the Mn with two unpaired electrons, so its moment should be $2.83 \mu_B$ (which differs slightly from the 2.5 for 20-250°C). The Me-Me bond may be broken at higher temperatures (the distance is very large), and localization of the electron on the Mn atom should raise the moment to $3.87 \mu_B$, which is in general agreement with our result.

Chemical crystallography leads us to expect that other monosilicides of the 3d metals with the $FeSi$ structure should also show an increase in effective magnetic moment with temperature.

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