

short note

reprinted from

**physica
status
solidi (b)**

phys. stat. sol. (b) 47, K99 (1971)

Subject classification: 18.4; 12.1; 21.1

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The Effect of High Pressure on the Néel Temperature of γ -Manganese

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γ -manganese is a metallic antiferromagnet with the Néel temperature of 540 °K, at which the crystal symmetry transforms from tetragonal to cubic with increasing temperature (1, 2). Since the pure γ -manganese is unstable at the Néel temperature, γ -manganese alloys with dilute concentration of other elements, such as the Mn-Cu system, have been studied extensively. The specific heats of Mn-Cu alloy have been measured over a wide range of Cu concentration (3 to 5).

It was reported that the entropies due to the latent heat and due to the gradual destruction of magnetic order with increasing temperature are 0.32 and 1.05 cal/mol deg, respectively, by extrapolation of these data to pure γ -manganese (5). The latter is about half the value expected from a localized model. The purpose of this study was to examine both magnetic and crystallographic transitions in γ -manganese alloys under high pressure and to correlate, if possible, the two phenomena. Differential thermal analyses under high pressure and specific heat measurements and thermal expansion measurements were made for a γ -Mn_{0.95}Cu_{0.05} alloy.

Samples were prepared from 99.9% pure manganese and electrolytic copper. Appropriate mixtures of these metals were melted in an induction furnace in an argon atmosphere, and were cast in an iron mold. The ingots were shaped into a cylinder (18 mm diameter, 25 mm long) for the specific heat measurement and rectangular prisms (2x2x3 mm³) for determination of the Néel temperature using the differential thermal analysis method under high pressure. Then the specimens were annealed at 1000 °C for 18 h in vacuum and were quenched in water.

The transition temperature of this alloy was observed under high pressure up to 40 kbar by using the differential thermal analysis method. The Kennedy type of piston cylinder high pressure apparatus was used for this experiment.

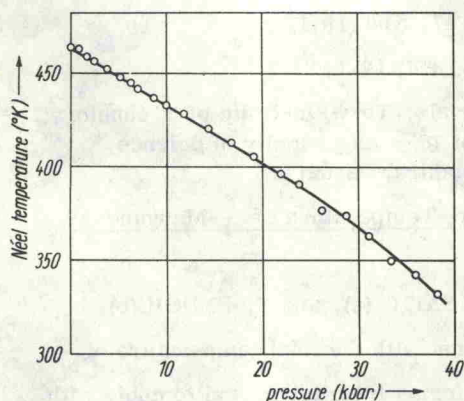


Fig. 1

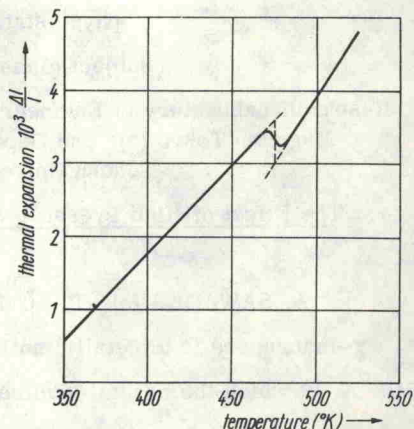


Fig. 2

Fig. 1. Pressure dependence of Néel temperature of γ -Mn_{0.95}Cu_{0.05} alloyFig. 2. Thermal expansion of γ -Mn_{0.95}Cu_{0.05} alloy

The specimen was covered with a gold foil, and placed in standard pressure cell fabricated from talc, graphite and pyrophyllite tubes containing AgCl powder to transfer pressure hydrostatically to the sample. The graphite tube was used as heater. The temperature was measured with an alumel-chromel thermocouple and no correction for the effect of pressure was made of the thermocouple e. m. f. The Néel temperature was obtained as a function of pressure as shown in Fig. 1. The variation is linear up to 25 kbar. The rate of decrease is $dT_N/dP = -3.25$ deg/kbar. The absolute value of the rate increases gradually with increasing pressure between 25 and 40 kbar.

The specific heat measurements were made from room temperature up to 500 °K in an argon atmosphere using an automatic adiabatic calorimeter. Three deep holes in the cylinders were used for the heater, the measuring thermocouples, and the temperature controlling thermocouples. A dc current was supplied to the heater, and the time required for the temperature to increase by 2.5 deg was counted automatically. The entropy change at the transition was estimated as $\Delta S = 0.16$ cal/mol deg.

The thermal expansion was measured from room temperature up to 500 °K using an unbonded strain gauge. The specimen was the same one as for the specific heat measurement. The dilatometer was calibrated using the thermal expansion of pure nickel. Results are shown in Fig. 2. The volume change at the Néel temperature was estimated as $\Delta V/V = 1.8 \times 10^{-3}$.

The entropy change, as calculated from the Clapeyron-Clausius equation, is given by $\Delta S = \Delta V(dP/dT_N)$. From the above data, $\Delta S = 0.099$ cal/mol deg. The value estimated from the specific heat measurement is 60% larger than this figure. This situation cannot be explained as an experimental error. Detailed studies are in progress.

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(Received August 26, 1971)