

Magnetization Measurements and Pressure Dependence of the Curie Point of the Phase Sc_3In †

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The approximate composition limits of the Sc_3In phase have been determined as 22 to 23 at. % In. Susceptibility measurements have been made on a number of alloys, containing various amounts of the Sc_3In phase, in the composition range 21-26 at. % In. Heat and mechanical treatments reveal a critical dependence of the magnetic properties on the degree of order of the Sc_3In phase. Although the susceptibility varies inversely with temperature, this behavior is not considered to be evidence for the presence of localized moments, since the magnetic moment per Sc atom is much smaller than that associated with $S = \frac{1}{2}$, and does not agree with the value obtained from the magnetization data as a function of magnetic field. Initial susceptibility measurements, below the Curie temperature, exhibit structure which suggests that a strong magnetocrystalline anisotropy may exist at temperatures close to T_c . The pressure dependence of the Curie point has been determined as $\partial T_c / \partial P = 1.9_8 \times 10^{-4} \text{ }^\circ\text{K bar}^{-1}$.

INTRODUCTION

THE theoretical understanding of magnetism in metals has, in the past,¹ been approached from the two extreme assumptions that the electrons responsible for the magnetic behavior are either situated in free- or nearly-free-electron energy bands (i.e., the itinerant model), or that they are located in discrete atomiclike orbital energy states (i.e., the localized model). The localized model has proved successful in accounting for the magnetic moments associated with the rare earths and their alloys (with the possible exception of cerium), but neither model, in its simplest form, has provided an adequate description of the magnetism of the transition metals and their compounds. This has led to a number of attempts to improve the agreement between experiment and theory by means of a suitable combination of appropriate features from both models. However, it is now¹ realized that the features which were considered to typify the localized, or Heisenberg model, can be reproduced in the itinerant model by the use of appropriate energy bands. Thus the important question which has to be answered in order to describe theoretically the magnetism of a particular system is to what degree does the spin-density distribution differ from that of a free-atom electron configuration. This question is of obvious interest when considering the reasons for the ferromagnetism of the compounds

ZrZn_2 ,² Sc_3In ,³ and Au_4V ,⁴ all of which have non magnetic metallic constituents.

Considerable effort has been devoted to the investigation of the magnetic properties of ZrZn_2 , both above and below its Curie point.⁵⁻⁸ Pickart *et al.*⁷ have interpreted their neutron diffraction data as showing that the spin density is predominantly associated with the zirconium atom, but more diffuse than that calculated from a $4d$ electron configuration. It was further observed that the spin density is anisotropically distributed and shows a marked pileup at the midpoint of the Zr-Zr bond. However, Foner⁹ has questioned this interpretation and has suggested an alternative explanation associated with the presence of magnetic impurities, which polarize their environment in a manner similar to the behavior of Fe impurities in Pd.^{10,11}

² B. T. Matthias and R. M. Bozorth, *Phys. Rev.* **109**, 604 (1958).

³ B. T. Matthias, A. M. Clogston, H. J. Williams, E. Corenzwit, and R. C. Sherwood, *Phys. Rev. Letters* **7**, 7 (1961).

⁴ L. Creveling, H. L. Luo, and G. S. Knapp, *Phys. Rev. Letters* **18**, 851 (1967).

⁵ S. C. Abrahams, *Z. Krist.* **112**, 427 (1959); S. Ogawa, *J. Phys. Soc. Japan* **20**, 2296 (1965); T. Yomadaya and M. Asanuma, *Phys. Rev. Letters* **15**, 695 (1965); C. E. Olsen, *J. Phys. Chem. Solids* **19**, 228 (1960); H. J. Blythe, *Phys. Letters* **21**, 144 (1966); F. E. Hoare and J. C. G. Wheeler, *ibid.* **23**, 402 (1966).

⁶ S. Ogawa and N. Sakamoto, *Phys. Letters* **23**, 199 (1966); *J. Phys. Soc. Japan* **22**, 1214 (1967).

⁷ S. J. Pickart, H. A. Alperin, G. Shirane, and R. Nathans, *Phys. Rev. Letters* **12**, 444 (1964); and in *Proceedings of the International Conference in Magnetism, Nottingham, 1964* (The Institute of Physics and The Physical Society, London, 1964), p. 223.

⁸ R. L. Falge, Jr., and R. A. Hein, *Phys. Rev.* **148**, 940 (1966).

⁹ S. Foner, *Bull. Am. Phys. Soc.* **12**, 311 (1967), and private communication.

¹⁰ A. M. Clogston, B. T. Matthias, M. Peter, H. J. Williams, E. Corenzwit, and R. C. Sherwood, *Phys. Rev.* **125**, 541 (1962).

¹¹ G. G. Low and T. M. Holden, *Proc. Phys. Soc. (London)* **89**, 119 (1966).

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¹ For general reviews see C. Herring, *Magnetism* (Academic Press Inc., New York, 1966), Vol. IV; and E. D. Thompson, *Advan. Phys.* **14**, 213 (1965).

TABLE I. Impurity content (ppm atomic) of starting materials and a Sc₃In phase alloy.

Impurity	ppm atomic		23.2 at.% In alloy ^b
	Scandium ^a	Indium ^a	
Al	33	5	40
Ag	2	1	30
Ca	11	6	4
Cu	35	1	9
Cd			20
F			<1
Fe	250	8	700
Mg	2	2	55
Na	10		4
Ni			15
Pb			10
Si	8	3	2
Sn			45
Th			3
Rare earths	N.D.	N.D.	130
			15
			1
			<100

^a Typical analyses as supplied by Johnson Matthey. Two lots of scandium were used in the preparation of the alloys.

^b Mass spectrographic analysis by Fulmer Research Institute.

Relatively little is, as yet, known about the magnetic properties of the other two compounds beyond the fact that they become ferromagnets at low temperatures. It is the main purpose of the present paper to report measurements of the magnetization of the Sc₃In phase as a function of magnetic field up to 40 kOe in the temperature range 1.25 to 32°K and measurements of the pressure dependence of the Curie point.

SAMPLE PREPARATION

Matthias *et al.*³ originally reported that the ferromagnetism of the Sc-In system occurs over the very narrow composition range from 23.2 to 24.2 at. % In. Compton and Matthias¹² showed that a superlattice structure exists in this composition range and from NMR measurements Wyluda *et al.*¹³ suggested that the ferromagnetism of the system was associated with this ordered phase. Accordingly alloys containing 21.1, 23.2, 24.2, and 26.2 at. % In were prepared by melting the constituents together in an argon arc furnace. These quoted compositions are those obtained after correcting for the weight loss which occurred during preparation, assuming that this was entirely due to the evaporation of In. The analysis of the starting materials, as given by the manufacturers is given in Table I.

Magnetic measurements and metallographic examinations were carried out on samples cut from these alloys in the "as cast" state, and following the heat treatment indicated in Table II. Metallographic examination of the "as cast" alloys revealed the presence of dendrites of a second phase which oxidized rapidly in air and this

phase has been tentatively identified as Sc₂In. Only in the case of the 21.1% alloy were we able to remove this phase completely by the anneal for one week at 800°C, although the amount present in the other samples was reduced. However, a new second phase was present in the annealed 21.1% sample, which was shown by x-ray examination to have the Sc structure with a lattice spacing close to that of scandium and it was therefore assumed to be a Sc-In solid solution phase. The relative volumes of the Sc₃In and Sc₂In present in the samples were determined using a Quantitative Television Microscope. A summary of the sample phase composition is given in Table II. Crude determinations of the liquidus temperature were made at 23.2 and 33.3% In and from these and the metallographic examinations a schematic phase diagram has been constructed which is presented in Fig. 1. The Sc₃In phase field is estimated to extend from approximately 22 to 23 at. % In at 800°C with no observed change in these limits down to 400°C.

Following the 800°C anneal the polished surface of the 24.2 at. % sample was examined by the glancing angle x-ray technique and superlattice lines were clearly resolved, the intensities of which were consistent with 100% order in the DO₁₉ hexagonal structure. Lattice parameters were obtained from filings (300 mesh), which had been given a further 4-day anneal at 400°C, using Cu K α radiation in an x-ray powder camera. The values were $a=6.56\pm 0.01$ Å; $c=5.12\pm 0.01$ Å, which may be compared with $a=6.421\pm 0.005$ Å; and $c=5.183\pm 0.005$ Å obtained by Compton and Matthias.¹²

Magnetic measurements were also made on a sample of the 23.2% In alloy, which had been annealed at

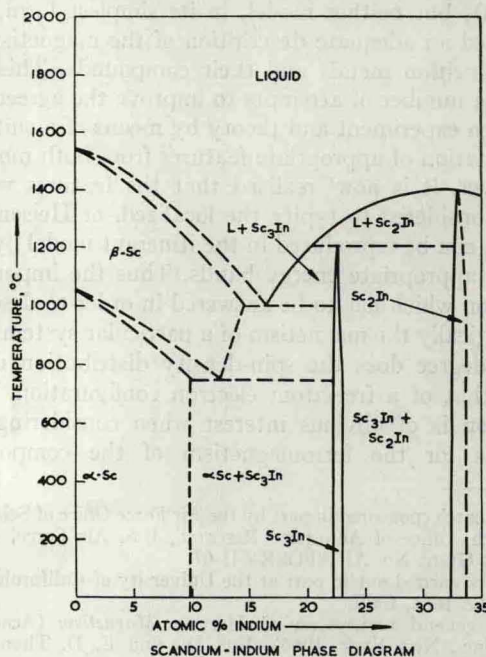


FIG. 1. Schematic phase diagram for the Sc-In system.

¹² V. B. Compton and B. T. Matthias, *Acta Cryst.* 15, 94 (1962)

¹³ B. J. Wyluda, R. G. Shulman, B. T. Matthias, and E. Corenzwit, *Phys. Rev.* 137, A1856 (1965).