Anomalous behavior of the weak itinerant ferromagnet Sc_3In under hydrostatic pressure

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The magnetic properties of three homogeneous samples of the weak itinerant ferromagnet Sc_3In have been determined in the hydrostatic pressure, temperature, and magnetic field ranges 0–6 kbar, 3–300 K, and 0–57 kOe, respectively. The pressure dependence of the Curie temperature $T_c(P)$ was determined to 31 kbar. The application of hydrostatic pressure is found to significantly enhance the magnetic properties of Sc_3In at both low and high temperatures relative to T_c. These results contradict a theoretical model by Wohlfarth, according to which the magnetic properties of homogeneous weak itinerant ferromagnets should be weakened under pressure.

I. INTRODUCTION

An adequate description of the many forms of magnetism in solids, in particular in metals, continues to pose a challenge to theoretical solid-state physics. Whereas magnetism in insulating solids can be normally described in terms of the behavior of an ensemble of local magnetic moments whose magnitudes can be determined by envoking Hund's rules and the appropriate crystalline-field splittings, in magnetically concentrated metals only ions of the rare-earth or heavy-actinide series are able to exhibit magnetism with local-moment character.[1] Magnetism in metals containing primarily the 3*d*-series elements Cr, Mn, Fe, Co, and Ni is generally believed to arise from exchange splitting in the 3*d* band. A general description of the conditions under which such itinerant electron or band magnetism can occur is a knotty theoretical problem which was first treated by Slater[2] and Stoner.[3] For many years the results of their model have provided a convenient theoretical framework for a qualitative description of magnetism in transition-metal compounds. A particularly simple limit of band magnetism is obtained for the so-called weak itinerant ferromagnets.[4] Because of the extremely small band splitting in the weak itinerant ferromagnetic state, the low-temperature high-field moment M_0 takes on a value of only a fraction of a Bohr magneton per transition-metal ion; the high-temperature Curie-Weiss moment p_{eff} can be an order of magnitude or more higher. Weak itinerant ferromagnets are also characterized by low values of the magnetic ordering temperature T_C. The weakness of the magnetism occurs because the sizable overlap of the magnetic orbitals in such systems places them near the magnetic-nonmagnetic transition.[1] Small perturbations, such as those caused by change of composition or application of pressure or magnetic field, can result in an appreciable change in the magnetic state. Since the application of pressure generally increases the overlap of the magnetic orbitals, one would anticipate that sufficient pressure would drive a weak itinerant ferromagnet into a nonmagnetic state. This expectation is supported by the following approximate expression proposed by Wohlfarth[5–7] for the pressure dependence of the Curie temperature T_C for an homogeneous ferromagnet:

$$\frac{\partial \ln T_C}{\partial P} = -\alpha / T_C^2 + \tfrac{5}{3}\kappa \; , \tag{1}$$

where the second term on the right-hand side originates from Lang and Ehrenreich,[8] α is a slowly varying quantity,[5] and κ is the compressibility. Since for weak itinerant ferromagnets both α and κ are positive and the first term in Eq. (1) dominates over the second,[5] the Curie temperature would be expected to *decrease* with pressure; in particular, the magnitude of $\partial \ln T_c / \partial P$ should be particularly large for systems with low values of T_C. These and other predictions of the Wohlfarth theory have received considerable support by the results of a series of experiments on ferromagnetic Ni-Pt alloys[9] and $Ce_{1-x}Y_xFe_2$ and Y-Fe compounds,[10] to a name a few. The success of Eq. (1) has led Wohlfarth to even suggest[5] that, should a given system show a pressure dependence $T_C(P)$ at odds with Eq. (1), this can be taken as a sign that the sample is inhomogeneous—a kind of theoretical microprobe. Indeed, Wagner and Wohlfarth[11] have shown that for heterogeneous ferromagnets

$$\partial \ln T_C / \partial P \approx -a + bT_C \ .$$

It should, however, be emphasized that a number of rather drastic approximations had to be made to arrive at the simple expression given in Eq. (1). In Refs. 5–7 Wohlfarth has pointed out that in the derivation of the above expressions the energy-band structure is assumed pressure independent: the Fermi energy E_F, the effective degeneracy temperature T_d, and the density of states $N(E_F)$ scale smoothly with some power of the bandwidth W which itself depends on the sample volume as $W \propto V^{-5/3}$ for a d-band metal.

Matthias *et al.*[12] have suggested that the most unequivocable examples of itinerant-electron magnetism occur in systems, like $ZrZn_2$, $TiBe_{2-x}Cu_x$, and Sc_3In, in which none of the constituents is ever known to exhibit local-moment paramagnetism in any metal. Whereas the Curie temperature for the first two compounds has been conclusively shown to decrease rapidly with pressure,[13] in agreement with Eq. (1), T_C is reported to *increase* with pressure for the single sample of Sc_3In studied.[14] Wohlfarth[5,15] has cited this latter result as evidence that the Sc_3In sample is inhomogeneous. In view of this unexpected experimental result for $T_C(P)$, and the apparently anomalous values of the lattice parameters reported in Ref. 14 for Sc_3In (see below), it would seem prudent to repeat the above experiment of Gardner *et al.* by applying hydrostatic pressure to a number of carefully prepared Sc_3In compounds.

In the present experiments we determine the dependence of the magnetic properties of Sc_3In on hydrostatic pressure to 6 kbar for three well-characterized homogeneous samples with varying In concentration; in a separate experiment, $T_C(P)$ is determined to 31 kbar. The present experiments confirm the anomalous increase of T_C under pressure found by Gardner *et al.*;[14] in addition, we find that for Sc_3In the low-temperature high-field magnetization M_0 and the high-temperature effective moment p_{eff} also increase with pressure. The present experimental results would thus appear to lie outside the region of validity of Wohlfarth's description of pressure effects in weak itinerant magnets.

II. EXPERIMENT

Three polycrystalline Sc_3In samples with compositions 24.1, 24.3, and 24.4 at. % In were prepared by one of the authors (K.I.) at the Ames Laboratory from 99.999% pure In and 99.9% pure Sc; full details on the sample preparation, as well as on the results of heat-capacity studies on the same samples, are given in a previous publication.[16] Chemical analyses of the scandium starting materials[16] revealed that the Sc used in the 24.3% In sample was of significantly higher purity than that used in the other two samples (9 ppm versus 53 ppm Fe impurity). The samples were first arc melted together and then sealed in a tantalum capsule. To promote homogenization and establish the ordered DO_{19} structure (Mg_3Cd type), the encapsuled samples were first annealed at 800 °C for 5 d and then at 700 °C for an additional 5 d before being allowed to cool at a rate of 0.1 °C/min to room temperature. Metallographic and electron microprobe analyses showed that all Sc_3In samples were to $\sim 1\ \mu$ resolution homogeneous single-phase compounds. X-ray diffraction experiments confirmed the Mg_3Cd-type structure for the three samples studied with the following values of the room-temperature lattice parameters: $a = (6.4302 \pm 0.0005)$ Å and $c = (5.1870 \pm 0.0006)$ Å, independent of the In concentration. These values are in quite good agreement with those published by Compton and Matthias,[17] who found $a = (6.421 \pm 0.005)$ Å and $c = (5.183 \pm 0.005)$ Å, but differ substantially from values quoted by Gardner *et al.*[14] $a = (6.56 \pm 0.01)$ Å and $c = (5.12 \pm 0.01)$ Å. It is conceivable that this difference could have a bearing on the pressure dependence of the magnetic properties.

Static magnetization studies over the range of temperature 3–300 K and magnetic field 0–7 T were carried out using a standard Faraday magnetometer which utilizes a Cahn R-100 microbalance and a superconducting solenoid with separate main and gradient windings. Measurements in fields as low as 16 Oe were possible by energizing only the gradient coil and positioning the sample near its center. For measurements at ambient pressure the Sc_3In sample was placed in a gold-plated 4.5-g crucible made from high-purity 99.999% ASARCO copper which contained less than 2-ppm magnetic impurity concentration. The diamagnetism of the Cu crucible was compensated to $\sim 1\%$ by a 0.49-g piece of Mo which had been purged of Fe impurities by repeated arc melting. All samples had ~ 100-mg mass and were shaped into right-angled parallelepipeds with approximate dimensions $2 \times 3 \times 3\ \text{mm}^3$; because of the weak magnetization of Sc_3In, it is not necessary to correct for demagnetization effects.

Low-temperature magnetization studies on Sc_3In under hydrostatic pressures to 6 kbar were carried out in the Faraday magnetometer by employing a magnetically compensated high-purity binary Cu-Be pressure clamp with 50-g mass. A thin piece of Pb was included in the pressure cell to serve as a superconducting manometer at low temperature.[18] The high-pressure Faraday magnetometer is described in more detail in previous publications.[19–21]

Measurements of the ac susceptibility of Sc_3In to hydrostatic pressures as high as 35 kbar were made possible by employing a metal-gasket cell with WC anvils which was originally developed by Fasol and Schilling[22,23] and adapted to ac susceptibility measurements by Eiling and Schilling.[18,24] In this pressure cell a small piece of the Sc_3In sample is placed together with a Pb manometer in the 0.3-mm-diam bore of a miniature primary-secondary coil system with outer diameter 1.0-mm and 0.8-mm height; the primary ac field was approximately 15 Oe at 33 Hz. In this work two additional leads were connected to the primary windings to allow a four-point measurement of the resistivity $\rho(T)$ of the primary winding. Since the resistivity of copper decreases rapidly under pressure [$\Delta R/R = -1.95 \times 10^{-3} P\,(\text{kbar}) + 5.25 \times 10^{-6} P^2$ at room temperature],[25] a measurement of the resistivity of the copper windings can be used to determine the pressure in the pressure cell. To extend the useful temperature range of the Cu manometer, we are currently measuring $\rho(T,P)$ for copper below ambient temperatures.[26] Experimental details of the metal-gasket pressure cell and the ac susceptibility measurement are described elsewhere.[18,22–24,27]

III. RESULTS AT AMBIENT PRESSURE

To lay the groundwork for an analysis of the high-pressure experiments, we first compare the results of our measurements of the magnetic properties of Sc_3In at ambient pressure to those of previous studies. In Fig. 1 the magnetic susceptibility at 57 kOe is plotted versus temperature for the 24.1 at. % In sample. Analyzing the susceptibility using a Curie-Weiss law plus a constant term

$$\chi(T)=C/(T-\Theta)+\chi_0 \qquad (2)$$

results in a linear χ^{-1} versus T plot if we set $\chi_0 = +110\times10^{-6}$ emu/mole (mole of $Sc_{0.759}In_{0.241}$). The data from the 24.3 at. % In and 24.4 at. % In samples can be successfully analyzed using the same value of χ_0.[27] From these analyses we list in Table I the derived values of the paramagnetic Curie temperature Θ and the effective moment per Sc atom $p_{\text{eff}}=(3kC/N\mu_B^2)^{1/2}$, where C is the Curie constant, N is the number of Sc atoms per mole of Sc_3In, k is the Boltzmann constant, and μ_B is the Bohr magneton. The agreement with the previous studies is seen to be reasonably good. The somewhat enhanced values of p_{eff} from Matthias *et al.*[28] presumably are a consequence of their setting $\chi_0=0$ in the above equation for $\chi(T)$. That the present value of $p_{\text{eff}}\simeq0.7$ lies well below that ($p_{\text{eff}}=1.73$) for $S=\frac{1}{2}$ is consistent with the proposed itinerant-electron nature of the magnetism in Sc_3In. The small positive values of Θ signal a possible ferromagnetic state at lower temperatures. The negative value of Θ for the nominal 24.4 at. % In sample of Matthias *et al.*,[28] as opposed to the positive value we find, presumably reflects differences in sample preparation between the two groups. The same authors[28] have, in fact, shown that in Sc_3In ferromagnetism only occurs within an extremely narrow concentration window ($\Delta x\simeq0.01$) about the concentration $x\simeq0.24$ in $Sc_{1-x}In_x$; the value of x depends itself on the thermal history.

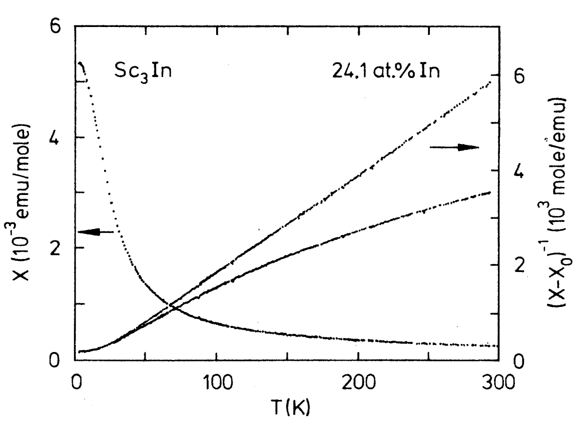

FIG. 1. Magnetic susceptibility of 57 kOe for Sc_3In with 24.1 at. % In and reciprocal susceptibility for $\chi_0=0$ and $\chi_0\neq0$ (see the text) as function of temperature. Units are electromagnetic units per mole of $Sc_{0.759}In_{0.241}$.

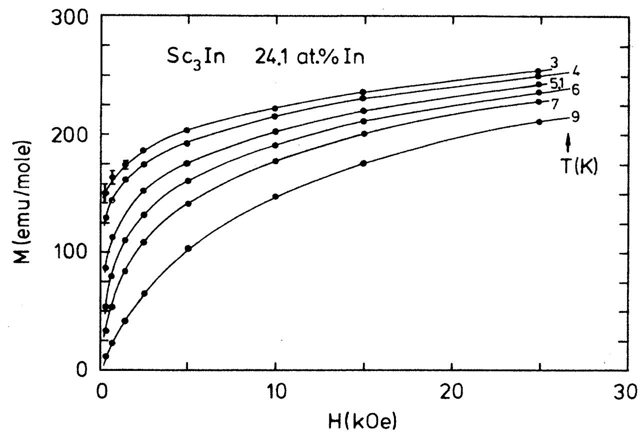

FIG. 2. Magnetization vs field for Sc_3In with 24.1 at. % In at various temperatures.

Magnetization measurements for the 24.1 at. % In sample to 25 kOe at various temperatures between 3 and 9 K are shown in Fig. 2. At 3 K an applied field of 25 kOe is not sufficient to saturate the magnetization; a further measurement on the 24.3 at. % In sample to 57 kOe gives a similar result. At 3 K and 57 kOe the magnetic moment per Sc atom for this sample is only $0.068\mu_B$, in good agreement with the value $0.072\mu_B$ extrapolated to these conditions from the magnetization data of Gardner *et al.*[14] (see Table I). The high-field susceptibility χ_{HF} of Sc_3In is finite and increases with temperature, extrapolating to the value $\chi_{\text{HF}}(0\text{ K})\simeq+1000\times10^{-6}$ emu/mole at absolute zero. In agreement with previous work, the present magnetization studies thus confirm that Sc_3In

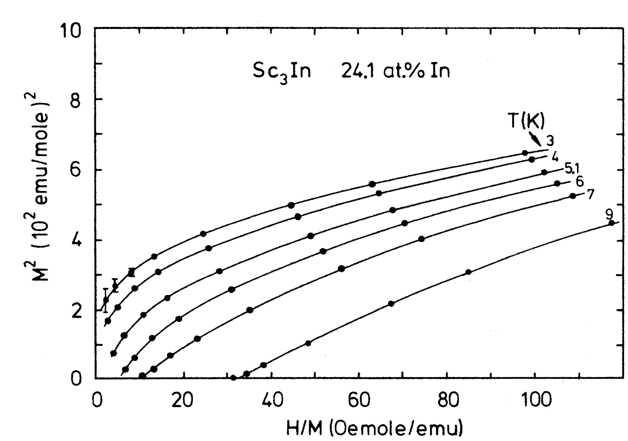

FIG. 3. Arrott plot for Sc_3In with 24.1 at. % In from magnetization measurements to 35 kOe.

does indeed exhibit the main characteristics of weak itinerant ferromagnetism, namely, low values of T_c, p_{eff}, and M_0, a large ratio of the magnetic moment derived from high-temperature Curie-Weiss to low-temperature high-field behavior, and the inability of high magnetic fields to saturate $M(T,H)$, even at temperatures well below T_C. Below we will discuss whether or not the Curie temperature and the other magnetic properties exhibit the strong decrease under pressure predicted by the Wohlfarth theory.

According to the Edwards-Wohlfarth theory[4] of weak itinerant ferromagnetism, the magnetization squared $M^2(T,H)$ should exhibit a linear dependence on $H/M(T,H)$. Such an "Arrott plot" is shown in Fig. 3. The isotherms are seen to deviate strongly from linear behavior, particularly for low values of field and temperature; similar deviations from linearity have been recently observed by Dhar *et al.*[29] on the itinerant ferromagnet Ni_3Al. This unfortunate circumstance precludes an accurate determination of the spontaneous magnetization for $H \rightarrow 0$; the Curie temperature T_C can also not be accurately estimated since it is not possible to ascertain from the data in Fig. 3 which isotherm passes through the origin. Similar results are obtained for the other two samples.[27] Precise data to lower values of the magnetic field (<5 kOe) than can be reliably set in the present Faraday magnetometer are necessary. Previous magnetization and specific-heat studies[16] in fields below 1 kOe by one of us (K.I.) allow the estimate that $T_c = 5.5$, 6.0, and 6.3 K for the 24.1, 24.3, and 24.4 at. % In samples, respectively, as listed in Table I; values of $T_C \simeq 6$ K for Sc_3In were also obtained in specific-heat studies of other groups.[30,31] Improved linearity in the Arrott plots is obtained if $M^{2.5}$ is plotted versus $(H/M)^{0.5}$. From such "modified" Arrott plots we can estimate from the present data that $T_C \simeq 4.2$, 4.6, and 4.7 K, respectively, for the above samples; these values lie approximately 1 K too low. Mohn *et al.*[32] have shown that spin fluctuations can lead to a sizable negative curvature in the Arrott plots of weak itinerant ferromagnets. An analysis[27] of the present data using this model[32] leads to the estimate $T_C \simeq 5.5 \pm 0.7$, 5.9 ± 0.2, and 6.3 ± 0.7 K, respectively, in excellent agreement with the low-field magnetization and specific-heat estimates of T_c discussed above. Spin-fluctuation effects have been shown[16] to have a sizable effect on the low-temperature specific heat of Sc_3In.

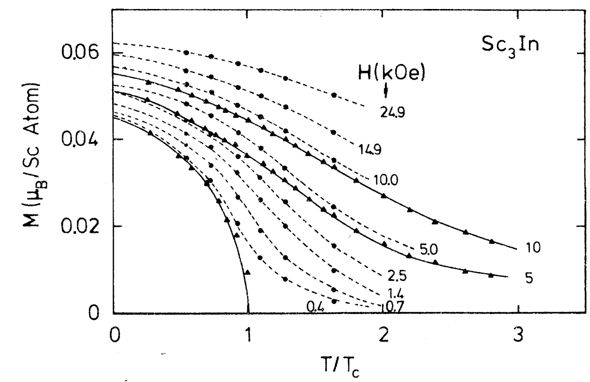

FIG. 4. Magnetization of Sc_3In vs relative temperature T/T_C for various values of applied field. Solid line from numerical calculation (Ref. 30), (▲) data points from Ref. 30 on 24.25 at. % In sample, (●) data points from present data on 24.1 at. % In sample, dashed line through latter data, points for clarity.

In the so-called unified spin-fluctuation or self-consistent renormalization theory (SCR) of band magnetism, Moriya[33] and others have developed a theoretical framework which contains elements of both the classical local-moment and Stoner-Wohlfarth itinerant-electron models. Within the SCR model the spin density can be well localized but fluctuates with time in both direction and magnitude. Takeuchi and Masuda[30] have applied this theory to a discussion of the magnetic properties of a Sc_3In sample with 24.25 at. % In, as shown in Fig. 4.

TABLE I. Applied field H, effective moment p_{eff}, paramagnetic Curie temperature Θ, Curie temperature T_C, and low-temperature high-field moment M_0 for present Sc_3In compounds compared to previous results by Gardner *et al.* (Ref. 14) and Matthias *et al.* (Ref. 28). Values for T_C are determined from Arrott plots at low fields (<1 kOe). Values of M_0 are followed by temperature and field (...K,...kOe) at which measurement was carried out.

Sample (at. % In)	H (kOe)	p_{eff} (per Sc atom)	Θ (K)	T_C (K)	M_0 (μ_B/Sc atom)	Reference
24.1	57	0.71	+11	5.5	0.068(3,57)	present
24.1					0.050(3,20)	present
24.3	57	0.70	+12.6	6.0	0.049(3,20)	present
24.3	10	0.69	+16.1			present
24.4	57	0.69	+11.9	6.3	0.050(3,20)	present
24.2	13	0.67	+16	7.5	0.051(4,10)	14
24.2					0.065(4,40)	14
24.2					0.056(1.2,10)	14
24.2					0.066(1.2,40)	14
24.0	14	0.81	+6	6	0.047(1.4,14)	28
24.1	14	0.80	+6	6	0.051(1.4,14)	28
24.2	14	0.75	+7	6	0.050(1.4,14)	28
24.4	14	0.72	−6			28

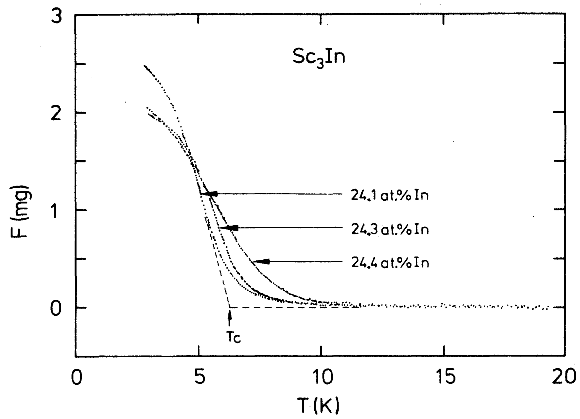

FIG. 5. Force change measured by Faraday balance as function of temperature for three Sc_3In samples in 16-Oe magnetic field. The construction leading to the estimate of T_C is shown.

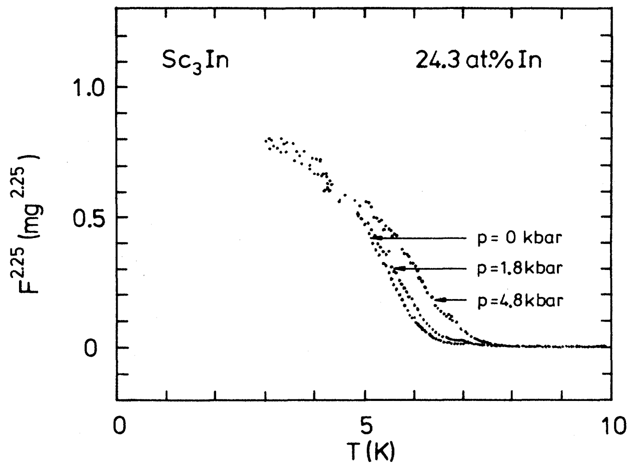

FIG. 6. Force change measured by balance to $\frac{9}{4}$th power vs temperature for 24.3 at. % In sample at three different values of the applied pressure. The Curie temperature T_C shifts rapidly to higher temperature under pressure.

The agreement between theory and experiment is remarkably good, allowing an estimate of $T_c = 5.5$ K for the Curie temperature which agrees with the low-field measurement[16] on our 24.1 at. % In sample (see Table I). In Fig. 4 the present data on our 24.1% In sample is included; the agreement between the two sets of isotherms is quite reasonable.

A further method of estimating the Curie temperature is to place the sample in a small field (here 16 Oe) and measure the initial susceptibility as the temperature is decreased. The magnetic ordering temperature is then estimated by the intersection point of the two dashed lines, one through the background and the other tangent to the inflection point, as seen in Fig. 5. The values of T_C so obtained lie $\sim 1-2$ K above those listed in Table I.[27] If we plot the susceptibility raised to the power α versus T, a different set of values for T_C results. If we arbitrarily set $\alpha = \frac{9}{4}$, the estimated values of T_C for the above three samples become 5.5, 6.1, and 6.3 K, respectively, in good agreement with the values from Arrott plots given in Table I.

IV. RESULTS AT HIGH PRESSURE

We would now like to discuss the results of the present high-pressure experiments in reference to the expectation of Wohlfarth that the magnetic properties, and in particular the Curie temperature, of a weak band magnet should decrease rapidly if pressure is applied. In Fig. 6 the onset temperature of the initial susceptibility is seen to shift rapidly to higher temperatures with increasing hydrostatic pressure. The values of T_C determined in the above manner from the data on the two Sc_3In samples measured are shown in Fig. 7. T_C is seen to increase approximately linearly with pressure at the rate

$$dT_C/dP \simeq +0.15 \pm 0.01 \text{ K/kbar}$$

$$(\text{or } d\ln T_C/dP \simeq +2.7\%/\text{kbar})$$

for 24.1 at. % In and

$$dT_C/dP \simeq +0.195 \pm 0.01 \text{ K/kbar}$$

$$(\text{or } d\ln T_C/dP \simeq +3.25\%/\text{kbar})$$

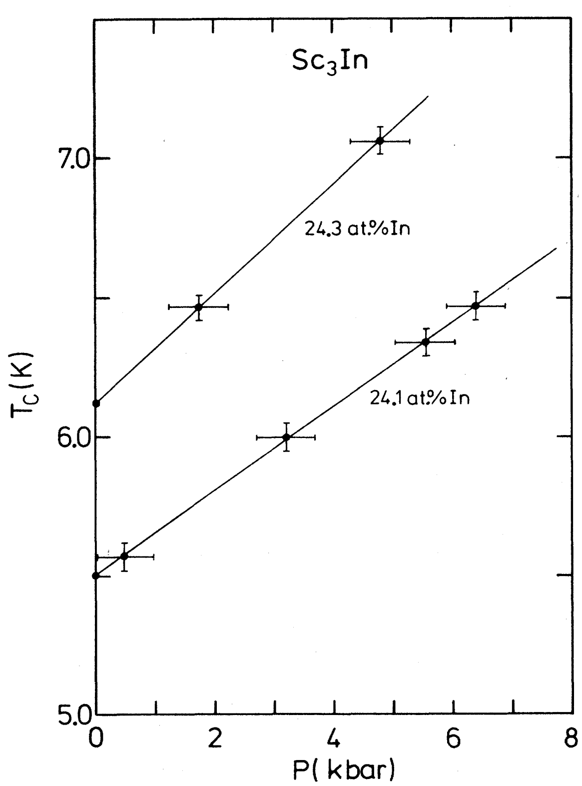

FIG. 7. Pressure dependence of Curie temperature T_C for 24.1 and 24.3 at. % In samples. Solid straight lines are drawn through data points with error bars for clarity.

TABLE II. Results of present high-pressure experiments. The relative change under pressure of the Curie temperature T_C, the magnetization M_0 at 3 K, and the effective moment p_{eff} at 40 K for the three Sc_3In samples studied.

Sample (at. % In)	P range (kbar)	$d\ln T_C/dP$ (%/kbar)	$d\ln M_0/dP$ (%/kbar)	$d\ln p_{eff}/dP$ (%/kbar)
24.1	0–5	+2.7	+0.85	
24.3	0–5	+3.25	+0.94	+0.6
24.3	0–31	+2.63		
24.4	0–5		+0.87	+0.5

for 24.3 at. % In; $d\ln T_C/dP$ thus appears to be slightly larger for the sample with higher In content. These results, which are summarized in Table II, are in good agreement with the results of Gardner *et al.*[14] to 14 kbar (+0.20±0.01 K/kbar) for a 24.2 at. % In sample. The data to 5 kbar on the 24.3 at. % In sample were extended to pressures above 30 kbar by use of the hydrostatic metal-gasket cell described above. In Fig. 8 the ac susceptibility curve signaling the onset to ferromagnetism is seen to shift to higher temperatures with pressure, yielding a linear $T_C(P)$ dependence with

$$dT_C/dP \simeq (+0.16\pm0.01)\ \mathrm{K/kbar}\ .$$

The sizable positive pressure derivative of T_C first measured by Gardner *et al.*[14] is thus clearly established as an intrinsic property of Sc_3In.

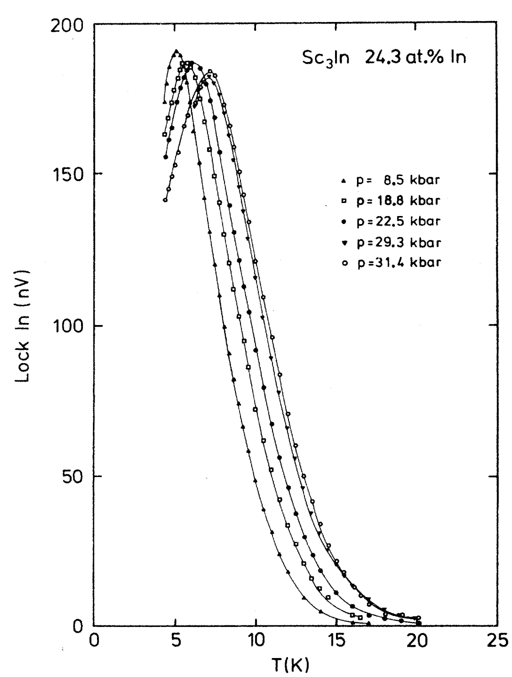

FIG. 8. ac susceptibility of 24.3 at. % In sample in ~15-Oe field vs temperature at hydrostatic pressures between 8.5 and 31.4 kbar. Solid lines are guide to eye.

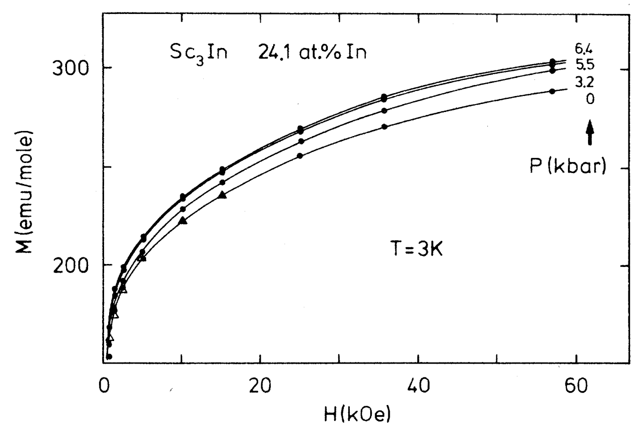

FIG. 9. Magnetization of 24.1 at. % In sample at 3 K vs field for various applied pressures to 6.4 kbar. Solid lines are guide to eye. The magnetization increases with pressure.

In the first high-pressure experiment on the 24.4 at. % In sample the sample was inadvertently squeezed between the two pistons of the pressure clamp, leading to a permanent deformation of ~4%. After removing the sample from the pressure cell, the initial susceptibility curve $\chi(T)$ at ambient pressure broadened markedly and the magnetization curve $M(H)$ at 3 K was reduced for all values of field H [at 1.5 and 25 kOe $M(H)$ was reduced by ~8 and 3.5%, respectively]. Gardner *et al.*[14] also reported that the magnetic susceptibility of Sc_3In is sensitive to heat treatment and mechanical cold work.

The effect of hydrostatic pressure on the magnetization curves $M(H)$ at 3 K for three Sc_3In samples was also studied. As is seen in Fig. 9, $M(H)$ *increases* with pressure at all values of the applied field; in fact, the *relative* change of $M(H)$ with pressure is found to be independent of the value of the field H. As seen in Table II, the relative change of the magnetization at 3 K under pressure $d\ln M_0/dP$ is only approximately one-third that of the Curie temperature $d\ln T_C/dP$. Both derivatives appear to be somewhat larger at higher In concentrations. The intermediate value of $d\ln M/dP$ for the 24.4 at. % In sample may originate from the previous plastic deformation of the sample. Averaged over the three samples measured, the relative change of M under pressure at 3 K is given by

$$d\ln M_0/dP \simeq (+0.86\pm0.03)\%/\mathrm{kbar}\ .$$

For comparison with theory the pressure derivative at 0 K is needed. From the measured pressure dependence of M at 3 K and $T_C(P)$, and the ambient-pressure dependence of M on the relative temperature T/T_C, it is possible to estimate[27] that within experimental error

$$d\ln M(0\ \mathrm{K})/dP = d\ln M(3\ \mathrm{K})/dP\ .$$

After clearly establishing that pressure enhances the low-temperature ferromagnetic properties of Sc_3In, it is of interest to ask what effect pressure has on the high-temperature paramagnetic state. Because of the relative weakness of the paramagnetic signal, the experimental errors inherent in this determination are relatively large. An analysis of the high-temperature susceptibility in terms of a Curie-Weiss law [Eq. (2)] allowed only an estimate of the upper limit $|d\ \ln p_{\text{eff}}/dP| < 2\%$/kbar. Higher precision can be achieved by repeated measurements at a single temperature. Analyzing the pressure dependence of the susceptibility at 40 K in terms of Eq. (2), assuming Θ and χ_0 pressure independent, allows the estimate

$$d\ \ln p_{\text{eff}}/dP \simeq (+0.6 \pm 0.2)\%/\text{kbar} .$$

In contrast to the expectation of the Wohlfarth theory of weak itinerant ferromagnets, we thus find that the application of pressure *strengthens* the magnetism of Sc_3In both in the paramagnetic and ferromagnetic states.

V. DISCUSSION

The main result of the present paper is that the magnetic properties of Sc_3In ($T_C, p_{\text{eff}}\ M_0$) are all significantly enhanced under the application of high hydrostatic pressures. That the magnetic properties change rapidly in unison was also observed by Matthias *et al.*[28] in studies across the $Sc_{1-x}In_x$ compound series, where $T_C, p_{\text{eff}}\ M_O$, and Θ were all observed to pass through a sharp maximum within a very narrow range ($\Delta x \simeq 0.01$) of $x \simeq 0.24$. Such a "movement in concert" supports the contention that Sc_3In is a weak itinerant ferromagnet, since in a local-moment ferromagnet p_{eff} and M_0 would normally remain constant. Although the present experimental accuracy was not sufficient to determine the pressure dependence of Θ, on the basis of the above we would anticipate that Θ would increase with pressure. If the present experiments were extended to higher pressures, the magnetic properties would be expected to eventually pass through maximal values and fall to zero. The present experiments almost double the pressure range of previous studies[14] and show that $T_C(P)$ continues to increase linearly with pressure to at least 31 kbar. That even higher pressures will ultimately weaken and destroy all forms of magnetism is anticipated on very general grounds.[1] Magnetism thrives best for well-localized atomic states where Hund's rules apply. Under certain conditions itinerant electrons can also exhibit magnetism, as in the case of weak itinerant ferromagnets. Under pressure well-localized electron states progressively delocalize and broaden into bands. If such an electron band is able to support ferromagnetism by splitting into spin subbands at low tempreatures, this magnetism will surely be lost at higher pressures where the demagnetizing effects from increasing bandwidth W dominate over magnetizing effects from increasing exchange interaction I. This latter point is well illustrated for jellium by a simple derivation by Bloch[34] where he shows that for a sample with volume V, $I \propto V^{-1/3}$ and $N(E_F) \propto W^{-1} \propto V^{+2/3}$ so that the quantity $IN(E_F) \propto V^{+1/3}$ *decreases* under pressure. The Stoner criterion for ferromagnetism $[IN(E_F) > 1]$ is thus only fulfilled for sufficiently low pressures.

Equation (1) is notably unsuccessful in accounting for the present experimental results on Sc_3In. For weak itinerant ferromagnets with low values of T_C, as in the present case, the first term in Eq. (1) ($-\alpha/T_C^2$), which is always negative, dominates[5-7] over the second term $5\kappa/3$, which is always positive. The value of T_C should, therefore, *decrease* under pressure. Even if we were to arbitrarily set the first term equal to zero, the remaining positive pressure dependence from the second term

$$d\ \ln T_C/dP = +5\kappa/3 = +0.38\%/\text{kbar}$$

(Ref. 35) is nearly an order of magnitude too small to account for the present results in Table II. Wohlfarth's conjecture that the samples studied by Gardner *et al.*[14] were strongly heterogeneous, rendering Eq. (1) inappropriate, would also seem to be incorrect; using samples whose homogeneity has been confirmed by microprobe analysis, we obtain an almost identical pressure dependence $T_C(P)$ as that in Ref. 14. We are thus led to conclude that the assumptions made in deriving Eq. (1) are too far reaching and simplified to allow a correct account of the pressure dependence of T_C for Sc_3In. Specifically, we suggest that Wohlfarth's assumption[5-7] that the band structure remains unchanged under pressure, and that $N(E_F)$ and T_d scale with the bandwidth W, may be inappropriate here. Equation (1) thus cannot account for changes in magnetic state brought about by a pressure-induced shift of one band relative to another which would change the relative number of electrons in each band. Detailed spin-polarized band-structure calculations are clearly essential for exploring these possibilities and obtaining a deeper understanding of the magnetic properties of Sc_3In at both ambient and high pressures. Equation (1) should, we believe, be regarded as a rough first-approximation result which may be useful in describing experimental trends in selected systems. However, one should not forget that the assumptions behind the simple expression for $T_C(P)$ in Eq. (1) may render it inappropriate for a particular system. We note that very recent studies[36] of the pressure dependence of the de Haas-van Alphen effect in the weak itinerant ferromagnet $ZrZn_2$ have revealed clear deviations from the Stoner-Wohlfarth model.

Wohlfarth[5] has derived the following simple expression relating the pressure dependence of M_0 to that of T_C:

$$\frac{d\ \ln M_0}{dP} = \frac{d\ \ln T_C}{dP} - 5\kappa/6 - \frac{d\ \ln B}{2dP} . \tag{3}$$

This relation was, in turn, derived by taking the pressure derivative of the more general expression

$$M_0^2 = T_C^2/[2N(E_F)\mu_B^2 T_d^2 B] \tag{4}$$

under the simplifying assumption used above that under pressure both $N(E_F)$ and T_d scale with the bandwidth W. B is given to second order in temperature as $B = 1/(2\chi_0 M_0^2)$, where χ_0 is the ferromagnetic susceptibility at 0 K. Wohlfarth[5] has contended that the last two terms on the right-hand side of Eq. (3) are generally small for weak itinerant ferromagnets; indeed, $-5\kappa/6 \simeq -0.2\%$/kbar is small compared to $d\ \ln T_c/dP \simeq +3\%$/kbar from Table II. Why, then, is the relative change of M_0 under pressure only about one-third as large as that of T_C? This apparent "discrepancy" may arise from the above assumption that $N(E_F)$ and T_d scale with W. If we consider the more general expression in Eq. (4), we see that a rapid increase of $N(E_F)$ with pressure would help lead to an equality between the pressure dependence of both sides of Eq. (4). Such a rapid increase in $N(E_F)$ could be caused, for example, by a pressure-induced change in the number of d electrons which could cause the Fermi energy to shift into a peak in the density of states.

In the above discussion it has been implicitly assumed that Sc_3In possesses a single spin lattice. The presence of two or more opposing spin sublattices, as in a ferrimagnet, would allow the possibility that the total magnetization could *increase* under pressure even if the magnetization of all spin sublattices were to *decrease*. Experiments employing polarized neutron diffraction should be carried out to investigate this possibility.

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