FIG. 2. A typical self-inductance–vs-temperature plot for the $x = 0.9$ solid solution. $L(T)$ is the temperature-dependent inductance, L_{max} is the maximum value of $L(T)$, and L_0 is the minimum value of $L(T)$.

served differences in the magnetic transitions or chemical composition if the samples were quenched in air or were slowly furnace cooled. Chemical analysis of these materials indicated they were stoichiometric to within 4 at. % and the ratio of As to Sb was within 1 at. % of the nominal value. Powder x-ray-diffraction patterns indicated the presence of MnO in some of the solid solutions. The presence of MnO should not affect the magnetic transition temperatures of these materials.

The self-inductance technique[13,14] was used to determined the FM-to-PM transition as a function of pressure and temperature. Hydrostatic pressure
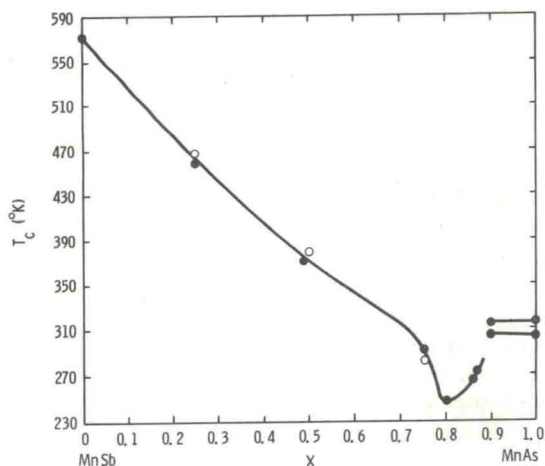
was applied with a 4.5-kbar helium-gas system[15] on the solid solutions which had transition temperatures less than 323 °K and with a Harwood 30-kbar liquid-pentane apparatus on the solid solutions which had transition temperatures greater than 323 °K. A typical reduced self-inductance–temperature plot as obtained for the $MnAs_{0.9}Sb_{0.1}$ solid solution is shown in Fig. 2. The transition temperature was arbitrarily taken as the half-transition point.

The experimental results are summarized in Figs. 3–5. In Fig. 3, the FM-to-PM transition temperature T_c is plotted as a function of concentration. The double curve in the concentration range $0.9 \leqslant x \leqslant 1.0$ is due to the thermal hysteresis associated with the first-order, hexagonal-FM-to-orthorhombic-PM, transition. No hysteresis is observed for the solid solutions in the concentration range $0 \leqslant x < 0.90$ which is indicative of a second-order FM-to-PM transition. Hereinafter we will refer to $0.9 \leqslant x \leqslant 1.0$ as the first-order region and to $0 \leqslant x < 0.9$ as the second-order region.

In Fig. 4, the initial pressure derivative of the FM-to-PM transition temperature $(\partial T_c / \partial P)$ is plotted as a function of concentration. The pressure derivatives were determined to within ± 0.15 °K/kbar. For MnSb, our measured pressure derivative of -3.0 °K/kbar is in good agreement with the value -3.2 °K/kbar as reported by Hirone et $al.$ [16] It is observed that $\partial T_c / \partial P$ changes almost precipitously in a very narrow concentration range ($\sim 3\%$) demarcating the first- and second-order regions. It should be remarked that the $x = 0.88$ material exhibited no thermal hysteresis at 4.5 kbar—indicating that the transition remained second order up
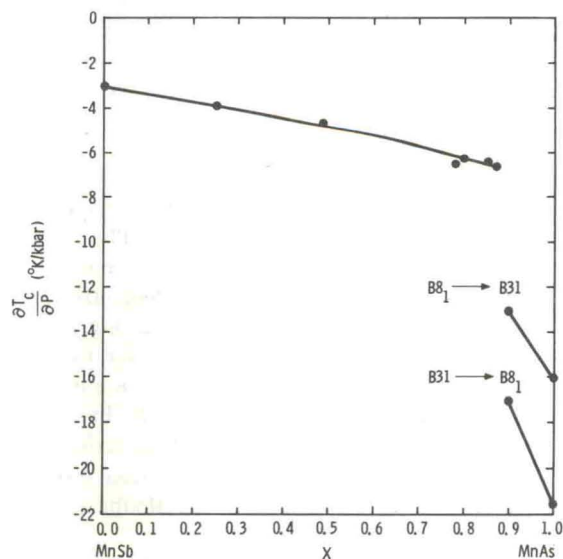

FIG. 3. Concentration dependence of the FM–to–PM transition temperature, in $MnAs_xSb_{1-x}$ solid solutions. Solid circle, present study; open circle, after Sirota and Vasilev (Ref. 4).

FIG. 4. Concentration dependence of the initial pressure derivative of the FM–to–PM transition temperature in $MnAs_xSb_{1-x}$ solid solutions.
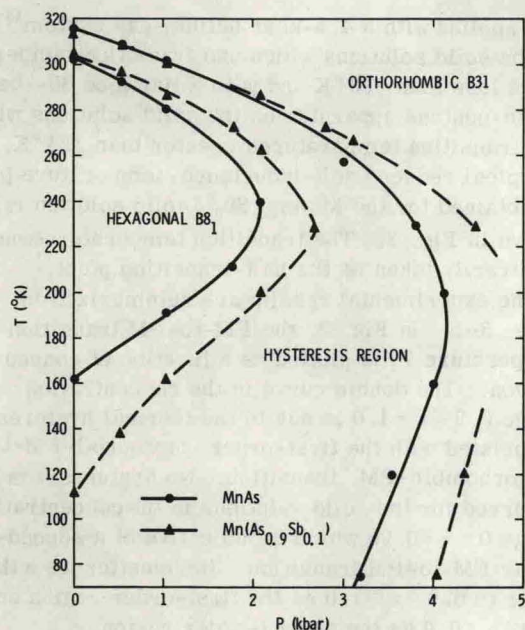
FIG. 5. Temperature-vs-pressure magnetic-phase diagram for MnAs and MnAs$_{0.9}$Sb$_{0.1}$.

to this pressure limit. (According to the Bean-Rodbell model,[17] it is possible that a second-order transition can be forced into a first-order transition under sufficient pressure; we shall comment more on this in Sec. III.

In Fig. 5, a portion of the temperature-pressure magnetic-phase diagram for MnAs and MnAs$_{0.9}$Sb$_{0.1}$ is shown. Our results for MnAs are in good agreement with the result of Menyuk et al.[1] It is observed, as speculated in Sec. I, that the substitution of 10% Sb for As does indeed increase the critical pressure required to stabilize the orthorhombic phase. The increase in critical pressure is approximately 0.75–1 kbar.

III. DISCUSSION

In Sec. III A, we discuss the solid solutions which exhibit second-order behavior. The results on these materials will be analyzed in terms of an itinerant-electron FM model. In Sec. III B, the alloys which exhibit a first-order behavior will be discussed in terms of the model proposed by Goodenough and Kafalas.[6] In addition, some comments will also be made on the Bean-Rodbell model[17] prediction of pressure-induced second-order to first-order behavior and on the equivalence of the itinerant-electron FM and the Bean-Rodbell models.

A. Second-Order Behavior

1. *Itinerant-Electron FM Model*

It is our purpose here to present an elementary

theory, unifying several existing theories, of a single-band itinerant-electron FM. In particular, we shall develop a theory, appropriate for 3d electrons, for the Curie temperature T_c and its pressure derivative $\partial T_c/\partial P$; and we shall show how estimates of the effective exchange I times the density of states at Fermi level $N(\epsilon_F)$ can be made from the measurements of $\partial T_c/\partial P$. The theory presented here follows quite closely the earlier work of Wohlfarth,[9] Edwards and Wohlfarth,[10] Shiga,[18] and Wohlfarth and Bartel,[12] but includes details which have not been discussed in these earlier works.

For our model we assume that the exchange splitting is given by $nI\zeta$, where I is the effective intra-atomic exchange (accounting for the electron correlations) between the itinerant electrons, n is the number of d electrons per atom, and ζ is the relative magnetization per electron arising from single-particle excitations. In the Stoner theory, the exchange splitting is $2k_B\theta'\zeta$, where $k_B\theta'$ is the molecular-field-approximation interaction; thus $k_B\theta' = \frac{1}{2}nI$. The single-particle excitations are described by the Stoner equations,[9,10] and in the limit of $\zeta \to 0$ for $T \to T_c$ we have, using a Sommerfeld expansion, that T_c is given by[19]

$$T_c^2 = T_F^2(\overline{I} - 1)/\overline{I} \quad , \tag{1}$$

where

$$\overline{I} = IN(\epsilon_F) . \tag{2}$$

Here $N(\epsilon_F)$ is the density of states per atom per spin at the paramagnetic Fermi level, and T_F is the effective degeneracy temperature.[9,10] In order for the system to be FM, we have from Eq. (1) the Stoner criterion $\overline{I} \geq 1$. In the following discussion we shall make some assumptions as to the nature of I and $N(\epsilon_F)$.

In general, we assume that the effective intra-atomic exchange between the itinerant electrons I is a compositionally averaged constant in the case of the FM behavior of alloys. For the MnAs$_x$Sb$_{1-x}$ solid solutions considered in this paper, I is the effective exchange appropriate for the Mn atoms. The particular form we shall use for I has been discussed by previous authors[12,18,20,21] and is given here as

$$I = I_b(1 + \gamma I_b/W)^{-1} \quad , \tag{3}$$

where I_b is the bare interaction, W is the bandwidth, and γ is a constant. In addition, we assume that the number of magnetic electrons n remains constant,[22] consequently $N(\epsilon_F)$ can be written as[12,18]

$$N(\epsilon_F) = \beta/W \quad , \tag{4}$$

where β is another constant and is related to γ. It is implied that W and thus $N(\epsilon_F)$ scale uniformly (uniform-scaling assumption) under volume changes.