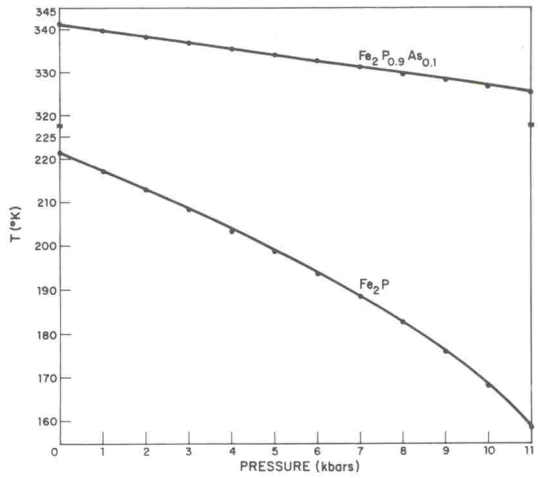

Fig. 1. Pressure dependence of the Curie temperature in Fe_2P and $Fe_2P_{0.9}As_{0.1}$.

using a gas generator with a vibrating-coil magnetometer, as described previously.[4] The Curie-temperatures T_c were obtained at low (100 Oe) fields, and plots of T_c vs hydrostatic pressure P are shown in Fig. 1. The points are experimental, and the solid lines are the analytic functions (with P in kbar and ΔT_c in °C)

$$P = -0.252(\Delta T_c) - 0.0012(\Delta T_c)^2 \text{ for } Fe_2P \quad (1)$$

$$P = -0.71(\Delta T_c) - 0.0017(\Delta T_c)^2 \text{ for } Fe_2P_{0.9}As_{0.1} \quad (2)$$

Extrapolation of eq. (1) to the extremum defined by $\partial P/\partial(\Delta T_c) = 0$ gives a critical pressure $P_c = 13.3$ kbar above which the ground state of Fe_2P should no longer be ferromagnetic.

Surprisingly, there was no significant change with pressure in the magnetization per molecule, μ, at 58K. Examination of μ vs the applied field H at T = 58K showed no appreciable change on passing from 1 atm to 10 kbar pressure. However, measurement of μ vs T/T_c for Fe_2P gave strikingly different results for 1 atm and 10 kbar. At H = 10 kOe, the inflection in μ vs T occurs at $T \approx 1.07\ T_c$ at P = 1 atm, but at $T \approx 1.15\ T_c$ at P = 10 kbar. Furthermore, an extraordinarily large exchange enhancement of the susceptibility, which extends to temperatures well above T_c, is markedly greater at 10 kbar than at 1 atm. At P = 10 kbar and $T = 1.02\ T_c$, a plot of μ vs H is extremely nonlinear, resembling the initial magnetization curve of a ferromagnet at $T < T_c$.

DISCUSSION

If the moment of Fe_2P were reduced from $\mu_o = 3.0\mu_B$ because of conduction-band overlap of E_F, pressure should change z_d, and hence μ_o,

more dramatically than T_c. Therefore, we conclude that the moment of Fe_2P is reduced because the molecular fields are not strong enough to empty all the antibonding states of antiparallel spin.

The existence of antiparallel-spin electrons in the ground state would create a ferromagnetic spin-density wave in the magnetically ordered phase (antiparallel-spin excited electrons create spin waves), and a spin-density wave reflects a long-range antiferromagnetic component to the interatomic-exchange interactions. Since longer M-M separations decrease the width of the 3d bands, the relative importance of this antiferromagnetic component must decrease with increasing As concentration x, which would account for the sharp rise with x in T_c (from 221 to 443K)[2] over the interval $0 \leqslant x \leqslant 0.33$. On the other hand, pressure would increase the antiferromagnetic component, and the critical pressure P_c presumably marks a transition from a ferromagnetic spin-density wave to a metamagnetic state.

The remarkable susceptibility above T_c in Fe_2P, and its enhancement by pressure, would seem to indicate that T_c is suppressed by pressure more rapidly than is the paramagnetic Curie temperature θ. Suppression of T_c relative to θ by weak, long-range antiferromagnetic interactions has been observed[5] in the metamagnetic thiospinel $Zn[Cr_2]S_4$, which contains localized 3d electrons. In Fe_2P, the ferromagnetic short-range order above T_c must be exceptional and appears to extend well above T_c, although the magnetic interactions are three-dimensional. This behavior is quite different from that found in $CoS_{2-x}Se_x$, where the ferromagnetic moment is also reduced because the bandwidth is too large.[6] The $CoS_{2-x}Se_x$ 3d bands are broadened with x, and the ferromagnetic-to-metamagnetic transition is marked by a reduction in θ that makes $\theta < T_c$.[7]

Within the molecular-field approximation, the paramagnetic Curie temperature θ is given by

$$\theta = (2/3k)S(S+1)\sum_v z_{uv} J_{uv} \qquad (3)$$

where z_{uv} is the number of v atoms near-neighbor to a u atom. If the interatomic exchange energy falls off more rapidly than linearly with decreasing atomic separation, then

$$J_{uv} \approx \sum_\ell J^o_{uv\ell} \sum_j \left[1 + \beta^j_{uv} \epsilon_j - \frac{1}{2} (\gamma^j_{uv} \epsilon_i)^2 + \cdots \right] \qquad (4)$$

The $J^o_{uv\ell}$ are components of the uv exchange interaction in the unstrained sample ($P = 0$) and $\beta^j_{uv} > 0$ because an $\epsilon_j < 0$ increases the 3d bandwidth, thereby lowering J_{uv}. The strain at equilibrium is given by[4]

$$\epsilon_j = \sum_i K_{ji} \left[\sum_{u,v} (\partial J_{uv} / \partial \epsilon_i) \vec{S}_u \cdot \vec{S}_v - P + T \sum_k \alpha_k c_{ki} \right] \qquad (5)$$

where K_{ji} = cofactor c_{ij}/(determinant c_{ij}), $\vec{S}_u$ and $\vec{S}_v$ are the thermodynamic expectation values of the spins at sites u and v, α_k is a thermal-expansion coefficient, and the c_{ki} are elastic constants. Since μ_o appears to be