to be small and negative where it is known. The sign of ΔH_3 agrees with the sign of ΔH_3 obtained for various impurities from the Mössbauer data of Stearns[3] (except in FeCr where ΔH_3 is negative). The positive sign of ΔH_3 and ΔH_4 also probably explains the average increase in the hyperfine field observed by Wertheim et al. in a Mössbauer experiment where the analysis yielded only ΔH_1 and ΔH_2 and the increase of the average hyperfine field with impurity concentration.[2]

Finally, we should like to emphasize the great advantage of using fast passage to examine the hyperfine field distribution. The modulation amplitude can be small and the detection can be performed at 90° to the modulation, which enables one to maintain the stable base line necessary when searching for weak satellites. We should also like to emphasize the necessity for examing low-concentration alloys ($c \sim 10^{-3}$) in order to resolve fourth- or fifth-nearest neighbors. A more complete paper including additional data will be published.

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MAGNETIC MOMENT OF Pd TO 150 kG: LIMITS OF EXCHANGE ENHANCEMENT IN Pd

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Magnetic moment data are presented for high-purity Pd at 4.2°K. The magnetic moment is a linear function of B up to 150 kG. On the basis of Wohlfarth's recent analysis of the high-field enhanced Pauli paramagnetism of Pd, the experiments determine limits of exchange enhancement for Pd and increase the minimum field for field-induced ferromagnetism in Pd.

The anomalously large static susceptibility χ in Pd is attributed to strong ferromagnetic exchange which enhances the Pauli paramagnetic spin susceptibility χ_P. The enhancement of χ is given by

$$\chi = \chi_P D, \qquad (1)$$

where $D = 1/[1 - N(0)V]$; $N(0)$ is the density of states at the Fermi surface, V is a parameter associated with an electron-electron interaction potential, and $\chi_P = 2\mu_B^2 N(0)$. Two aspects of exchange enhancement in Pd are of great interest: First, the effects of a large exchange enhancement lead to critical spin fluctuations which suppress superconductivity in Pd and Pd alloys and can also lead to enhancement of the electronic specific heat,[1,2] and second, there results a prediction of the transition in Pd from a strongly paramagnetic to an ordered ferromagnetic state at very

high fields (a lower limiting field H_C of 500 kG is estimated for occurrence of ferromagnetism by Wohlfarth and Rhodes).[3] The magnitude of both these effects depends critically on the value of D. Although estimates of D from 3 to 50 have been made,[1-10] experimental determinations of D have been very indirect so far. In this Letter we examine the high-field magnetic moment of Pd at 4.2°K in fields to 150 kG. We follow the discussion given by Wohlfarth,[5] who showed that for large D a noticeable field dependence in χ should be observed well below the estimated critical field H_C for ferromagnetism in Pd. Within the limits of our measurements we find that χ is independent of B. Our data are then used to place limits on the exchange enhancement factor D in a self-consistent manner which in turn increases the lower bound on H_C.

Wohlfarth[4] has shown that the field dependence of the susceptibility $\chi(B)$ is given by

$$\chi(B) = \chi(0)[1 + \tfrac{1}{2}\nu\mu_B{}^2 D^3 B^2] = \chi(0)[1 + \beta B^2], \quad (2)$$

where

$$\nu = \left[\frac{N''}{N} - 3\left(\frac{N'}{N}\right)^2\right]_{E=E_F}, \quad (3)$$

$$\chi(0) = \frac{n\zeta\mu_B}{B} = \frac{2N(0)\mu_B{}^2}{[1 - N(0)V]} = \chi_P D, \quad (4)$$

$$\chi(B) = n\mu_B \partial\zeta/\partial B. \quad (5)$$

Here the initial susceptibility at $B = 0$ and $T = 0$°K is given by $\chi(0)$, the differential susceptibility is given by $\chi(B)$, and the relative magnetization is $\zeta = (n_\uparrow - n_\downarrow)/n$, where $n_\uparrow$ and $n_\downarrow$ are the total number of electrons in the up- and down-spin bands per atom, and n is the total number of spins per atom. The quantity ν involves first (N') and second (N'') derivatives of the density of states $N(E)$ which are not yet evaluated with sufficient precision. To obtain Eq. (2), Wohlfarth assumes that the electron-electron interaction term V is a constant, that the relative magnetization ζ is small so that terms of higher order than ζ^3 can be neglected, that the density of states $N(0)$ at E_F is differentiable, and that the term βB^2 is much less than 1. The condition $\beta B^2 \ll 1$ is readily satisfied in our field range as demonstrated by the experiments discussed here.

The interesting features of Eq. (2) are these: (a) $\chi(B)$ is strongly dependent on B and D so that for sufficiently high B and D (and ν) a noticeable nonlinear $\chi(B)$ should be observable; (b) the sign and magnitude of ν depends on the band structure; and (c) the absolute value of $\chi(0)$ is not crucial—examination of the field dependence of χ is sufficient for our purposes. Because various estimates of D are quite high for Pd, high-field magnetic-moment measurements in Pd should yield useful boundaries for exchange enhancement and/or ν.

The magnetic-moment measurements were made in water-cooled high-power dc solenoids to 150 kG with three pure polycrystalline Pd materials. Samples composed of Johnson Matthey spectroscopic grade wires (with nominally 2 to 3 ppm Fe impurity) were examined. The measurements employed a very low-frequency vibrating-sample magnetometer,[11] where the sample moves between two axial series-opposing coils and each flux change is integrated and displayed on a recording versus time. Suitable low-pass and high-pass filtering is employed to reduce the background field fluctuations to an acceptable level and the resultant output is time-averaged to further increase the signal-to-noise ratio.

Results for these three samples are shown in Fig. 1, where the relative moment σ_n (normalized at 150 kG) vs B at 4.2°K is plotted. Sample III was a cylinder cast from Pd wire stock of the same batch employed for the Sample-II data. All the data closely fit a straight line over the entire field range. Since we measure magnetic moment rather than differential susceptibility, we integrate Eq. (2) to obtain

$$\sigma(B) = \int_0^B \chi(B)dB = \chi(0)[B + \tfrac{1}{6}\nu\mu_B{}^2 D^3 B^3] + \sigma_0$$
$$= a_1 B + a_3 B^3 + a_0, \quad (6)$$

where σ_0 is the spontaneous moment. Although σ_0 is expected to be negligible for pure Pd, the σ-vs-T data at low field show evidence[12] of some magnetic contribution even for 2 to 3 ppm Fe (since the Fe is effective in polarizing several hundred Pd atoms[13] with a resultant giant moment of about $13\mu_B$/Fe atom).

Least-squares computer determinations of the parameters in Eq. (6) were made in order to study any systematic changes with field. The results of various polynomial fits are tabulated in Table I for the three samples in Fig. 1. The general features are as follows: The smallest standard error (S.E.) in a_1 is usually observed for a strictly linear variation of σ (i.e.,

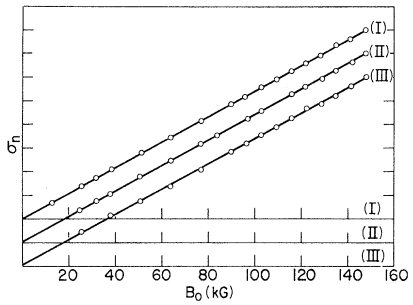

FIG. 1. Normalized magnetic moment σ_n versus applied field at 4.2°K for various Pd samples. (I) Johnson-Matthey (J & M) spectroscopic grade Pd (nominally 2 ppm Fe in Pd). (II) J & M (nominally 3 ppm Fe in Pd). (III) Arc melted cylinder of Pd fabricated from J & M wire of (II) above. All σ_n are normalized to 1.00 for the maximum value of B.

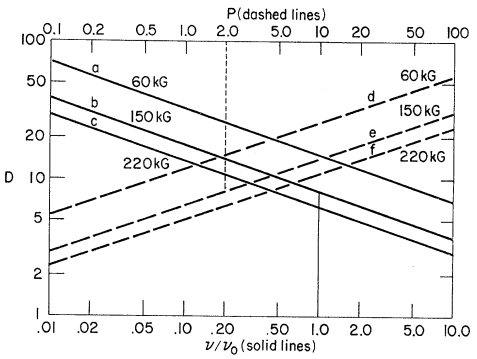

FIG. 2. Exchange enhancement D versus normalized band parameter ν/ν_0 (lower abscissa), and versus percent deviation from linearity, $P=100(\nu\mu_B{}^2D^3/6)B^2$ (upper abscissa). All axes are plotted on a log scale. Solid lines a, b, and c, corresponding to $B=60$, 150, and 220 kG, respectively, indicate limits set by present experimental resolution to 150 kG. Similarly, dashed lines d, e, and f, corresponding to 60, 150, and 200 kG, respectively, yield the limits of D vs P assuming $\nu/\nu_0=1$. The vertical line at $\nu/\nu_0=1$ to line b represents the present limits of D. The corresponding intercept for the P scale intersects line e.

$\sigma_1=a_1B$). In addition, the other polynomial fits σ_{10}, σ_{13}, σ_{130} (see Table I) yield values of a_1 and a_3 which are approximately equal to their respective standard errors. Furthermore, the results for the three samples are essentially the same within our experimental error. We conclude that the best fit is given by a strictly linear variation of σ with B. The values of of the cubic (a_3) and constant (a_0) term reflect our present experimental error. A very slight systematic decrease in a_1 is observed as a successively higher field points are added to the lowest field point, and the S. E. decreases monotonically. This is expected for saturation of trace amounts of Fe impurities in Pd.[12] Examination of a_3 as successively higher field points are added to the lowest field points, or as successively lower field points are added to the highest field points, shows that a_3 remains within the limits of experimental error. It should be noted that a_3 is apparently negative, partly reflecting possible saturation of trace localized moments at low fields. However, analysis of only the high-field points does not show any systematic positive value of a_3 for all the samples. In order to estimate limits of a_3 set by our experiments, we use a value of $|a_3|=0.65\times10^{-17}$ emu/g G^3 which reflects the limits of the experiments.

Although the effect of reduced D may be offset by increasing B in Eq. (6), present technical developments place a stringent upper limit on available B! Here we consider various estimates of ν and then establish limits of D. Wohlfarth[5] estimates an upper limit of $\nu=\nu_0$ [when $N'(E)=0$] of $\nu_0=1.18\times10^{26}$ erg^{-2}. This leads to a limit on D, based on the present data, of $D\leq8$ as indicated in Fig. 2. Studies of the Fermi surface and χ in Pd and in dilute Pd alloys are needed for more accurate

Table I. Polynomial fits of normalized magnetic moment σ_n versus Field B for three Pd samples in Fig. 1.[a]

Polynomial / Sample	$\sigma_1=a_1B$	$\sigma_{10}=a_1B+a_0$		$\sigma_{13}=a_1B+a_3B^3$		$\sigma_{130}=a_1B+a_3B^3+a_0$		
	a_1	a_1	a_0	a_1	a_3	a_1	a_3	a_0
I	6.74 (3.2)	6.52 (5.9)	2.1 (1.2)	6.82 (7.7)	−6.6 (5.5)	6.08 (15.)	+20. (6.2)	4.5 (1.0)
II	6.72 (1.9)	6.61 (3.6)	1.1 (0.9)	6.80 (4.8)	−6.0 (3.1)	6.51 (12.)	+ 4.3 (4.7)	1.5 (0.9)
III	6.81 (1.1)	6.76 (1.8)	0.5 (0.5)	6.90 (2.0)	−7.0 (1.4)	6.87 (4.3)	− 5.9 (2.0)	0.1 (0.4)

[a]The standard error for each coefficient is tabulated in the adjoining parentheses. Note different exponents for S.E. tabulations: Units, a_0 (S.E.) $=10^{-2}$ (10^{-2}) emu/g; a_1 (S.E.) $=10^{-6}$ (10^{-8}) emu/g G; a_3 (S.E.) $=10^{-18}$ (10^{-18}) emu/g G^3. Normalized $\sigma=\sigma_n=\sigma_1=\sigma_{10}=\sigma_{13}=\sigma_{130}=1$ emu/g at $B=150$ kG.

estimates of ν. For the present we indicate the range of ν and D in Fig. 2 based on our experiments up to 150 kG which place upper limits on $|a_3| = 0.65 \times 10^{-17}$ emu/g G^3 or $P \simeq 2$ at 150 kG. As mentioned above, this limit largely reflects the limits of experimental error rather than a real number. Since no accurate values of ν are yet available, Fig. 2 may be employed to test and/or eliminate theoretical predictions as they become available. Given an estimate[14] of D, an upper limit of ν is determined by our experiments; or conversely, given ν we have an upper limit of D consistent with the present data. Finally, given ν and D, the deviation from linearity, P, is given for characteristic values of B_0. It should be emphasized that the field dependence of σ suggests that β may be negative, whereas for a high-field ferromagnetic transition $\beta > 0$ is essential.

Figure 2 also shows the sensitivity of our estimates of D on both the range of B and the experimental resolution. Assuming that the field of dependence remains within our present experimental error ($P \simeq 2$) and $\nu/\nu_0 = 1$, the limits of D for 60 and 220 kG are indicated (Fig. 2, curves a and c). The 60 kG corresponds to that readily available with superconducting magnets, but it should be emphasized that large effects of trace amount of Fe or other localized moments must also be eliminated before such low-field data are meaningful. The value of 220 kG corresponds to a relatively large-volume dc field currently available at the National Magnet Laboratory. The effect of experimental resolution (or real deviations from a strictly linear σ vs B) is indicated by the dashed lines [Fig. 2, curves d, e, and f). Combining both of these results, it is apparent that at present a technically limited improvement is feasible with higher fields and possibly more improvement may be expected with higher resolution of the susceptibility measurements.

The most serious limitation is the lack of sufficiently precise values of $N(E)$ vs E. Because the limits on D vary as $\nu^{1/3}$, accurate theoretical values of ν to within factors of 2 or 3 would be quite useful. We expect that much more accurate estimates of ν will be forthcoming in the near future based on more detailed band calculations[15] and recent de Haas–van Alphen experiments[16] so that the present experiments may be applied to more accurate determinations of the exchange enhancement in Pd.

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