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Band-Structure Effects in the High-Field Magnetization of Pd and Dilute Pd–Rh and Pd–Ag Alloys*

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From Stoner theory we show that the magnetic field-vs-spin magnetization has the form $B(\sigma) = \sigma/\chi(0) + \Delta B(\sigma)$. The low-field susceptibility $\chi(0)$ is exchange-enhanced but, if the effective exchange potential does not depend on magnetization, the deviation from linearity ΔB may be derived solely from the band density of states $N(E)$. The previously reported Pd band-structure calculations, which were in excellent agreement with the de Haas–van Alphen data, have been extended to yield $\Delta B(\sigma)$ for Pd and, using the rigid band model, also for dilute Pd–Rh and Pd–Ag alloys. A van Hove singularity near the Fermi level of the 7% Rh–Pd alloy has a profound effect on ΔB. Our results are consistent with the measurements of Foner and McNiff, but detailed comparison is difficult since ΔB is of the same order as the experimental uncertainty; moreover, the experimental data have previously been analyzed according to a power series expansion of $\sigma(B)$ that neither separates the effect of band structure from that of exchange enhancement nor applies when van Hove singularities are important. Our formulation suggests a way of analyzing the experimental data.

Recent low-temperature measurements of the high-field magnetization of Pd [1] and dilute Pd(Ag) [2] and Pd(Rh) [3] alloys have been analyzed according to the formula

$$\sigma(B) = \chi(0)[B + (1/6)\nu\mu_B^2 D^3 B^3], \qquad (1)$$

which is the truncated power series in magnetic field B obtained from Stoner theory. Here σ is the magnetization; $\chi(0) = D\chi_P$ is the low-field susceptibility, which is exchange-enhanced above the Pauli spin susceptibility $\chi_P = \mu_B^2 N$ by the Stoner factor $D = (1 - NV/2)^{-1}$. The band density of states for both spins is $N \equiv N(E_F)$, and $\nu \equiv (N''/N) - 3(N'/N)^2$. It has been suggested that, once highly accurate band-structure calculations are available, the measurement of the high-field magnetization is well suited for the determination of D.[1] One reason is that the evaluation of $\nu^{1/3}$ is supposedly more accurate than the evaluation of N. From detailed relativistic APW calculations of the Pd band structure, we find that this is not the case.[4] Moreover, for those dilute alloys where deviation from linearity of σ vs B has been observed, we find that, within the rigid-band approximation, the density of states is not analytical over the relevant range and, consequently, Eq. (1) does not hold. In this paper we rewrite the Stoner–Wohlfarth result (1),[5] and give numerical results for the band-structure contribution to the nonlinearity in the curve of B vs σ.

Let the exchange interactions among the d holes be represented by the $\mathbf{k}$-independent potential V, and let spin–orbit coupling be neglected. In the presence of a magnetic field and the exchange interactions, the spin-up and the spin-down bands split rigidly by an energy 2ϵ. If this splitting is considered a function of magnetization m, the function $\epsilon(m)$ depends only on the integrated density of states $n(E)$, which is the number of electron states with energy less than E in the nonsplit bands. Specifically $\epsilon(m)$ is the odd part of a function $E(m)$, which is the inverse of $m(E) \equiv n(E) - n(E_F)$. The relation between field and magnetization is obtained by minimizing the total energy with respect to m :[5]

$$B = [\epsilon(m) - Vm/2]/\mu_B. \qquad (2)$$

If V is independent of magnetization, a deviation from linearity $\Delta B(\sigma)$ of the curve B vs σ, where $\sigma = \mu_B m$, is solely a band-structure effect, and we obtain

$$B = [\sigma/\chi(0)] + [\sigma/\chi_P]p(m). \qquad (3)$$

Here the leading term m/N of $\epsilon(m)$ is included in the linear term of Eq. (3), and $p(m) \equiv (N/m)\epsilon(m) - 1$. If $N(E)$ is analytical in the relevant range around E_F, $p(m)$ has as leading term $-(\nu/6N^2)m^2$. If the latter term is substituted into Eq. (3), and the resulting power series in $\mu_B m$ is inverted, we obtain $\sigma(B)$ as given by Eq. (1). However, Eqs. (2) and (3) yield a separation of band-structure and interaction effects, and the assumptions of a magnetization-independent V and an analytic $N(E)$ are not needed. Spin–orbit coupling may be approximately taken into account by writing the above formulas in terms of B, m, N, and V, and then replacing μ_B by $\mu_B g_{\text{eff}}/2$, where g_{eff} is an effective g factor.

We have found that the partial state density for the d-like holes in Pd (4th and 5th bands) contributes 90% to the total state density and is highly energy dependent,

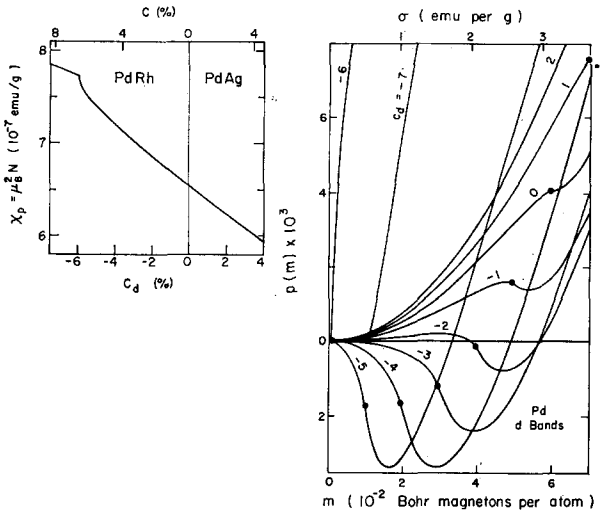

FIG. 1. The Pauli susceptibility $\mu_B^2 N$ and the function $p(m)$ appearing in Eq. (3) for Pd and, in the rigid-band approximation, for dilute $Pd_{1-c}Ag_c$ and $Pd_{1-c}Rh_c$ alloys. These results are derived from the Pd band-structure Eq. (4) for $g_{eff}=2$.

due to the presence of a saddle point in the purely d-like part of the 5th band just 1.89 mRy below the Fermi level.[4,6] The number of d states, computed by tracing of constant-energy surfaces $E_k=E$, is for the assumed potential given with a differential accuracy of 10^{-4} states/atom by[4]

$$n(E)=-433.3+34.66E-1.458E^{3/2}-0.615E^2, \quad (4)$$

which is valid for $-1<E<8$, and $E^{3/2}\equiv 0$ for $E\leq 0$. The number of states is in units of 10^{-3} states/atom, and energies are in mRy above the energy of the van Hove singularity.

From Eq. (4) we find for Pd,

$$\chi_P=6.55\times 10^{-7}(\text{emu/g})\cdot(g_{eff}/2)^2,$$

$$\nu=-0.15\times 10^{26}\ \text{erg}^{-2}.$$

Most recent de Haas–van Alphen data[7] seem to indicate that the error arising from the uncertainty in the crystal potential used in our *ab initio* calculation may approximately be compensated for by a shift in E_F of no more than -0.3 mRy relative to the d bands or, equivalently in the rigid-band language, by a shift of 1% towards Rh in nominal alloy composition.[4] The induced shifts in χ_P and ν are 2% and 25%, respectively. The extreme sensitivity of ν to the position of the Fermi level is due to the presence of the van Hove singularity. For the dual purpose of illustrating the uncertainty in our estimate of ν and of making predictions for dilute Pd(Rh) and Pd(Ag) alloys under the rigid-band assumption, we have calculated the band-structure contribution to $\Delta B(\sigma)$, expressed in terms of χ_P and $p(m)$, for various Fermi levels each corresponding to an addition of c_d electrons per atom to the pure Pd system. These results are shown in Fig. 1. Here the dots indicate the positions of the singularities, and it may be seen how the range of validity of a power expansion for $p(m)$ decreases as the Fermi level approaches the energy of the saddle point. For this position of the Fermi level, which corresponds to 6.6 at. % Rh in Pd, ν diverges, and the magnetization saturates rapidly: $\Delta B\approx 20$ kG $(\sigma/\text{emu/g})^{3/2}$. However, for the more dilute PdRh alloys, Pd and PdAg alloys the relative deviations from linearity of the theoretical B-vs-σ curve $Dp(m)$ do not exceed a few percent in the magnetization range obtainable.

From the experimental $\chi(0)$ data[1] for pure Pd we calculate $D=10.5$ when we take $g_{eff}=2$. Our high-field results for Pd and PdAg are consistent with the experiments,[1,2] since no systematic deviation from linearity larger than the experimental uncertainty of about 2% was observed for fields up to 150 kG, which corresponds to magnetizations up to approximately 1.0 emu/g. For $Pd_{0.95}Rh_{0.05}$, nonlinearity *was* observed,[3] and if we take $D=16$ and $g_{eff}=2$ our band theoretical curve passes through all the experimental points (B,σ) shown in Fig. 1 of Ref. 3. However, to get agreement with the theoretical upturn in the σ-vs-B curve for low magnetization, we had to assume that $\chi(0)$ was a few percent lower than that explicitly stated by Foner and McNiff.[3] This illustrates that a σ-vs-B plot is too crude and, since fitting the experimental data to a power law in B or σ does not make sense for this alloy, we suggest an experimental plot of B/σ vs σ be compared with our theoretical $p(\sigma/\mu_B)/\chi_P$. If the experimental points are accurate enough, useful conclusions about the assumptions of rigid bands and magnetization-independent exchange interactions might be possible.

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